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FACTORS INFLUENCING STRONTIUM-90 IN MILK from the BRAINERD, MINN. MILKSHED

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FACTORS INFLUENCING STRONTIUM-90 IN MILK from the BRAINERD, MINN. MILKSHED

by
Minnesota Department of Health
and
University of Minnesota

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U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service

Division of Radiological Health
Washington 25, D. C.

FOREWORD

National interest is focused on the extent to which radionuclides produced by nuclear detonations are found in our environment. The following report reflects this interest by analyzing factors which influence the strontium-90 concentrations in milk produced on selected farms in the Brainerd, Minnesota, milkshed.

Only through such studies as this will a firmer understanding be gained concerning the fate of radionuclides as they pass through the environment into man.

In view of the long and successful history of combined action by State and Federal agencies in promoting the Nation's health, it is appropriate that this first report from the Division of Radiological Health in the Environmental Health Series should represent such a concert of effort. The State of Minnesota Department of Health developed the results shown and discussed in Part I of this report, and the University of Minnesota was primarily responsible for the results reported in Part II. Both groups and the U. S. Department of Health, Education, and Welfare, through the Public Health Service and its Division of Radiological Health, jointly sponsored the study described in this report.

While the important contributions of the individuals involved in the Brainerd study are acknowledged on the following page, particular appreciation is extended to Dr. Robert N. Barr, Secretary and Executive Officer, State of Minnesota Department of Health, and his staff for their valuable liaison services on the project.

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ABSTRACT

A study was undertaken to investigate consistent farm-to-farm differences in the concentrations of strontium-90 found in milk produced within a small milkshed in central Minnesota. Covering an 8-month period, the study included sampling of milk, hay, grain, silage, water, forage, soil, and precipitation. The data indicate that 70 to 90 percent of the cattle's strontium-90 intake came from the native grass hay and that the farm-to-farm differences of strontium-90 concentrations in milk correspond to similar differences of strontium-90 concentrations in the hay fed. On the basis of the data obtained, it is not possible to explain why the consistent differences in the native grass hay existed.

FACTORS INFLUENCING STRONTIUM-90 IN MILK FROM THE BRAINERD, MINNESOTA, MILKSHED

INTRODUCTION

The purpose of this study was to obtain information concerning concentrations of strontium-90 present in the environment, particularly with relation to the factors contributing to its occurrence in milk and to determine, if possible, the cause or causes for observed differences in the concentrations of strontium-90 in the milk produced on selected farms.

Four dairy farms in the Brainerd, Minnesota, milkshed were chosen. The investigation covered a period of 8 months and included sampling of milk, cattle feeds and forage, water, soil, and precipitation. Since the total area involved was only 100 square miles, it was thought that geographic variability of fallout, including rainout, would not be significant. Similarly, general farming practices; age, size, and composition of the dairying herds; dairying practices; milk-handling techniques; and cattle-feeding routines were found, upon interview, to be surprisingly consistent from farm to farm. Thus a minimum of variation would be expected from these factors.

This project was the outgrowth of other radiation surveillance studies of the Minnesota Department of Health. In September 1958 the Department initiated a routine program of surveillance of the concentrations of strontium-90 in Minnesota market milk. Arrangements were made for the submission of monthly samples composed of daily portions of "grade A" milk from 6 bottling plants serving various parts of the State.[1] The samples were analyzed in the laboratories

[1] Original stations at Duluth, Minneapolis, Thief River Falls, Worthington, Brainerd, and Faribault. Subsequently, Rochester was substituted for the station at Faribault.

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of the Division of Environmental Sanitation, Department of Health, for strontium-90 and calcium concentrations. Early data from this program showed consistent differences of strontium-90 concentrations between sampling stations. The general magnitude of these differences persisted even though month-to-month results from all stations were variable and generally rising.

Speculation as to the causes of these differences led to the establishment of a number of precipitation collection stations in the spring of 1959.[2] Although data obtained from the analysis of the precipitation samples showed area-to-area variations, they did not account for the magnitude of the differences noted in the milk surveillance program. A comparison of the average concentration of strontium-90 in milk and the gross beta activity deposited by rain is shown in table 1. The milk data are expressed in units of micromicrocuries per liter and those for precipitation, in units of microcuries per square meter. During the period covered, the monthly samples from the Duluth and Brainerd milksheds showed strontium-90 concentrations approximately 2 to 4 times those obtained from other stations in the network.

Table 1.--Comparison of Strontium-90 in Milk and Total Year Old Beta Activity Deposited by Rain

Location	Average Sr^{90} activity in milk (μμc/liter) January-August 1959	Total one year old beta activity deposited by rain ($\mu c/m^2$) March-August 1959
Brainerd	20.8	[a] -
Duluth	20.7	1.7
Thief River Falls	13.8	1.5
Worthington	7.0	1.1 (Tracy)

[a] Precipitation collection station not located at Brainerd.

The Duluth milkshed consisted of over 600 individual suppliers, a substantial proportion of whom were located in Wisconsin, whereas the Brainerd milkshed consisted of only 16 producers. Both the Duluth and Brainerd milksheds were marginal farming areas. The Brainerd milkshed was selected for more detailed study because of the smaller number of suppliers involved.

In July 1959, each of the 16 farms of the Brainerd milkshed was visited and the owners were interviewed about practices relating to the dairying operations. It was found that the herd size averaged approximately 20. Eighty percent

[2] Duluth, Minneapolis, Thief River Falls, and Tracy.

of the dairy cows were Holstein. Milk production per farm averaged approximately 600 pounds per day with a farm-to-farm daily production range of 300 to 1300 pounds for that time of year. It was also learned that the farms were largely self-supporting; i.e., nearly all cattle feeds were grown on the respective farms. Dietary mineral and protein supplements were obtained from outside sources. When grain, hay, or fodder was purchased for one reason or another, the informants indicated that the materials were usually obtained locally. One-liter samples of milk for strontium-90 analysis were obtained from 11 of the farms during the July visit. Strontium-90 concentrations in micromicrocuries per liter found in these samples are shown in table 2.

Table 2.--Results of Preliminary Milk Samples from Brainerd Milkshed

Farm no.	Strontium-90 concentrations (μμc/liter)	
	Series I	Series II
1	27.3	22.0
2	15.0	15.8
3	16.3	17.3
5	19.0	21.0
7	24.4	18.3
8	21.5	15.6
10	15.2	15.8
12	32.4	23.3
14	28.6	21.3
15	16.4	11.9
16	27.5	26.5

Approximately 3 weeks after the July visit, an additional set of samples was obtained. Although the strontium-90 concentrations of the second series of "grab" samples were generally lower than the first set (average of 19 compared to 22 micromicrocuries per liter), the relative farm-to-farm concentrations in one series were generally comparable to those in the other series. It was apparent that within the Brainerd milkshed, which had demonstrated consistently higher than average strontium-90 concentrations compared to the State as a whole for the monthly composite program, there also existed a range of variability of concentrations on the basis of individual farms. This range of variability was about twofold, with the lowest concentrations being approximately equal to the average of the monthly surveillance program and the highest approximately twice the State average.

These preliminary findings were discussed with experts in the fields of soils, plants, and farming at the Institute

of Agriculture, University of Minnesota. The Brainerd project was proposed following these discussions. The study was initiated November 1, 1959, and terminated June 30, 1960. This project was financed in part by the Division of Radiological Health, U. S. Public Health Service. For administrative purposes, the prime contractual arrangements were made between the Minnesota Department of Health and the Public Health Service. A subcontract was then negotiated with the University of Minnesota for investigation of those phases of the study reported mainly in Part II of Results, Discussion, and Conclusions.

EXPERIMENTAL PROCEDURES

Material

GENERAL AREA - THE BRAINERD MILKSHED

Sketch maps of Minnesota and the Brainerd area (figure 1) show the location of the Brainerd milkshed. The surface mantle consists of glacial sediments deposited by the Patrician ice sheet and is mainly sandy and loose-textured, with many cobblestones and small boulders. (A detailed description of the profile of typical Brainerd soil is included in Appendix I, p. 51.)

The normal annual rainfall in this area is 25 inches with 17 inches falling during the growing season, which averages 131 days and extends from May 15 to September 24.

Farms in this area are organized with combinations of livestock (predominantly dairy cattle), grain, and hay enterprises. Hay, silage, and grain for the livestock are primarily home-grown, but some protein concentrates may be purchased for supplemental feeding. The size of the dairy herd is usually limited by the hay and small grain (mainly oats) production of the individual farm. The hay crop is generally a mixture of legumes and grasses (red clover and timothy with some alfalfa and brome grass). Corn is the principal crop for silage. The relatively short growing season limits the production of corn for grain.

SELECTED FARMS WITHIN THE BRAINERD MILKSHED

Separate farm units within the milkshed are indicated in the shaded areas of figure 2. The farms numbered 2, 10, 12, and 14 were selected for investigation after review of the data in table 2 and consideration of factors apparent at the time of the original visit and interview. Aerial photographs of the individual farm sites are included in the appendix, p. 52. A distribution for each of the selected farms is also included in the appendix, p. 56.

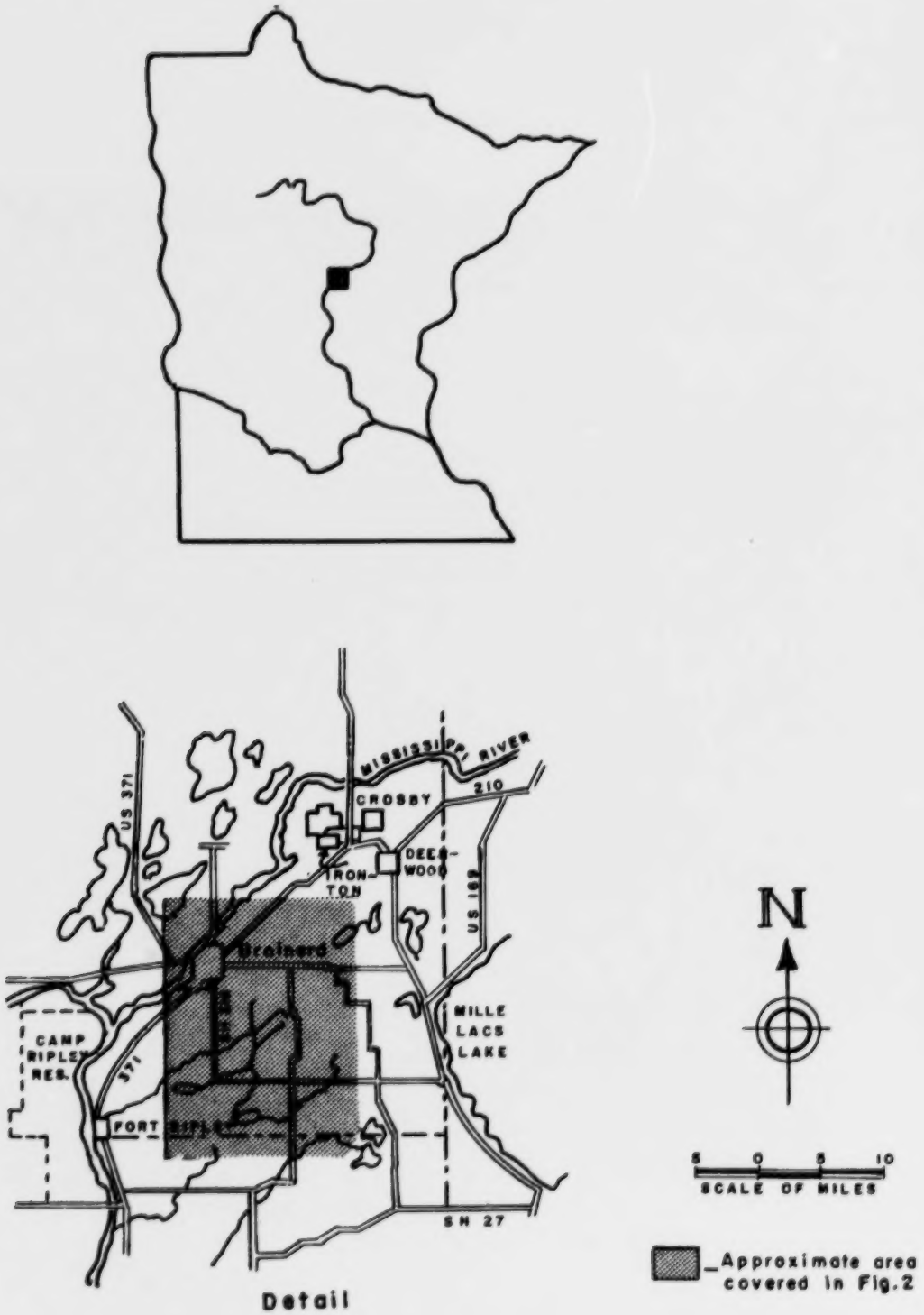

Figure 1.--Location of the Brainerd Milkshed

Sampling Methods

MILK

Milk samples were collected biweekly simultaneously with feed samples. They were taken from the bulk storage tank and therefore represented a total herd composite of from 1 to 4 milkings. All milk samples were analyzed for strontium-90, cesium-137, and calcium content.

CATTLE FEEDS

Cattle feeds were sampled biweekly during the survey. Generally, 3 types of feeds were collected: hay, grain and grain supplement combination, and corn silage. An estimate of the daily consumption of each feed was made by the farm operator at the time of sampling. During periods in which the cattle were feeding primarily on pasture, samples of the pasture growth were collected as part of the sampling. Feed samples were analyzed for strontium-90 and calcium, with selected samples being analyzed for total strontium also.

WATER

Well-water samples were collected biweekly, but no surface or ponded waters were found available for cattle consumption during the study. The water samples were analyzed for gross beta activity only since the concentrations found were not sufficient to warrant strontium-90 analysis.

Weekly precipitation samples were collected during most of the sampling period, including the entire spring rainy season. Collection stations were set up on each of the selected farms for this purpose. The stations consisted of a funnel with a 0.4 square meter collection area supported over a 12-gallon polyethylene pail. The samples were analyzed for strontium-90 content only.

SOILS

Soil samples were obtained at the start of the survey, November 2, 1959, and again at identical sites on May 18,

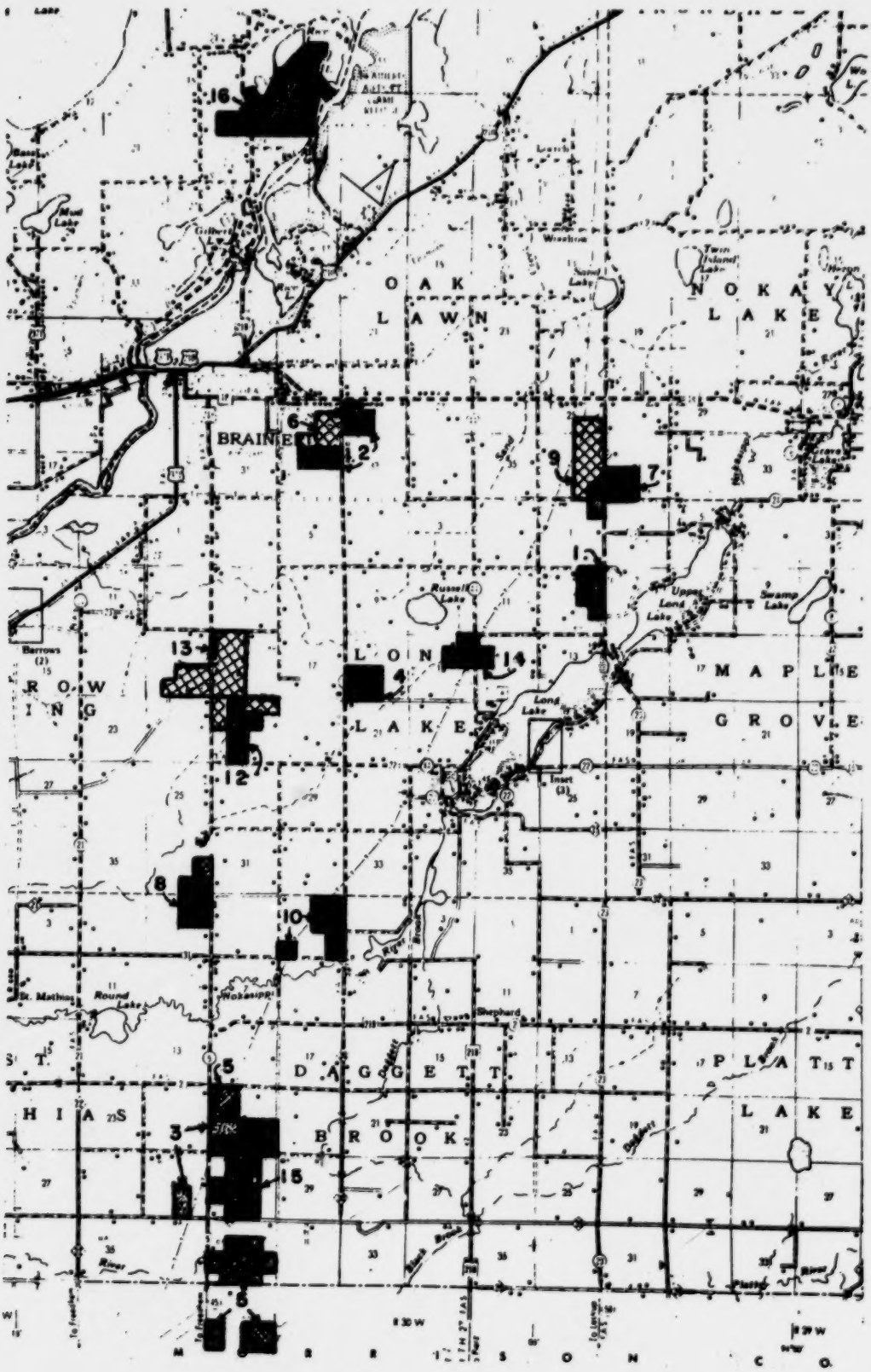

Figure 2.--Location of Farms

1960. Sampling sites were selected on the basis of topography, land use, and soil type. All locations, with one exception, were taken from land used for permanent pasture. Three sites (upland, lowland, and marsh) were selected from each farm unless an adequate representation of a given farm could be obtained by fewer samples.

Both surface and subsurface samples were obtained at each site and analyzed separately. Each consisted of a composite of 20 cores according to the method recommended by Hamada et al (6). A cylindrical sampling tool, 3 inches in diameter and 2 inches high, was used to obtain a surface sample to a depth of 2 inches. The subsurface samples (2-8 inches) were obtained with a bucket-type auger of the same diameter. Analyses included pH, percent organic matter, available phosphorous, exchangeable potassium, calcium and magnesium, total exchange capacity, total calcium and strontium, strontium-90, and cesium-137.

VEGETATION

Samples of vegetation were obtained from each soil sampling site at the time the soil samples were collected. These consisted of top cuttings obtained by using a sickle bar mower in the approximate area of the soil samples. The species of plants present in the cuttings were identified and their relative abundance was estimated at the time of sampling. In addition to these aerial vegetation samples, root mats, separated from surface soils, were also separately processed for analysis. The vegetation samples were analyzed for strontium-90 and calcium content.

Because of the possibility that other analyses of the samples might prove useful at some later date, representative specimens of all samples were stored for future reference.

Method of Analysis

MILK, CATTLE FEEDS, AND PRECIPITATION

The basic procedure used for the determination of strontium-90 as it applies to milk is explained in Appendix V,

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p. 64. Some modifications were made in the initial ashing or leaching steps for materials other than milk. In general, the sample size was selected so that it would contain approximately 1 gram of calcium, and 20 or 40 milligrams of stable strontium for carrier was added. The amount of carrier depended on the natural strontium content of the material being analyzed.

The method consisted of precipitation of the calcium and strontium as oxalates at a pH of 4. The oxalate was then destroyed with peroxide and nitric acid and the solution scavenged with yttrium and barium (as chromate) in the presence of ammonium chloride. Yttrium-90 was then allowed to ingrow in the presence of stable yttrium carrier and reprecipitated under the conditions prevailing in the scavenge procedure. The yttrium precipitate was then converted to the oxalate, filtered, and counted.

Chemical recovery of strontium was determined by flame photometry. During part of the study calcium was also determined by flame analysis. In these analyses lithium was used as an internal standard. The effects of various concentrations of calcium on the strontium-lithium response ratio were plotted. The analyses were performed using a recording photometer. On all other occasions the calcium was determined by titration with EDTA.

Both counter efficiency and over-all efficiencies for the various stages in the basic procedure were determined using a standard solution of strontium-90 - yttrium-90 (at equilibrium).

In addition, the procedure itself was checked against various samples of ashed material furnished by the Atomic Energy Commission, by analysis of pooled samples provided by the U. S. Public Health Service, and by analysis of wheat samples of known strontium-90 content supplied by General Mills, Inc.

Counts were repeated on about 25 percent of the samples to check the rate of decay of the separated yttrium.

For hay, silage, and precipitation samples, counting times were selected such that the counting error was never greater than 10 percent at the 95 percent confidence level. For milk and grain samples, the possible counting error at the 95 percent confidence level was somewhat greater for some of the very low-level samples. These counting errors

were estimated to be of about the same magnitude as the analytical error associated with the procedure.

The general procedure was originally developed for milk analysis, but was actually used more for other types of samples during this study. A shorter method was developed for the milk analysis after the start of the survey. In the shorter method, the milk samples were ashed in the usual way and yttrium carrier was added. When the samples were old enough to insure strontium-90 - yttrium-90 radio-equilibrium, the yttrium was precipitated as a phosphate at a pH of 2.6. Decay curves demonstrated the absence of other radioactive materials precipitating under these conditions for the milk samples. This shorter method was not suitable for other types of samples, however, and they were analyzed using the original procedure.

SOILS

The soil samples were air dried and surface organic matter was separated from the surface sample by hand for separate analysis. The soils were passed through a heavy duty ore grinder which reduced the particles to about 25 mesh size. After thorough mixing, subsamples were removed for laboratory analysis.

Exchange capacity was determined by the method of Holt (7), in which the exchangeable cations of the soil are replaced by silver. The silver is subsequently removed in the presence of excess thiourea and the excess thiourea is titrated amperometrically with standard silver solution.

Exchangeable calcium and magnesium determinations consisted of a soil extraction with ammonium acetate solution. The total exchangeable calcium plus magnesium was found by titration with EDTA. The calcium was determined by flame photometry and magnesium was thus determined by difference.

Total strontium determinations, as well as electrodialysis procedures used in this study, are relatively new and were developed in part to carry out the study. A detailed description of the electrodialysis procedure is included in Appendix V, p. 64. The procedure used for total strontium was developed independently by the Soils Department of the Institute of Agriculture and a detailed report of it is in press.

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RESULTS, DISCUSSION, AND CONCLUSIONS

This section has been divided into 2 parts since the data were developed under separate contractual arrangements. Part I presents the data developed by the Minnesota Department of Health, and Part II reports those phases of the study developed primarily by the University of Minnesota.

Part I

RESULTS

Milk

The data obtained from the analyses of the individual milk samples from each farm unit are shown in table 3. The table includes the date of sampling, the data in terms of micromicrocuries of strontium-90 per liter and grams of calcium per liter, and the calculated strontium-90 to calcium ratio. The farms with generally high strontium-90 concentrations in milk are indicated by the letter (H) after the farm number. In like manner, the low farms are designated by (L). The high and low farm designations used in subsequent sections of this report relate to the strontium-90 concentrations in the milk produced. Cesium-137 data for the individual milk samples are included in the appendix.

Although the dairy herds were in the barn for winter feeding during most of the sampling period, several of the samples collected in the spring were obtained after pasture feeding had started. The starting dates for spring pasture at each farm were as follows:

Farm 2(L)	-	May 23, 1960
Farm 10(L)	-	May 18, 1960
Farm 12(H)	-	May 9, 1960
Farm 14(H)	-	May 9, 1960

Data in tables 3, 4, 5, and 6 should be reviewed with these dates in mind.

Table 3.—Concentrations of Strontium-90 and Calcium Found in Milk

Date sampled		Farm 2(L)			Farm 10(L)			Farm 12(H)			Farm 14(H)		
		μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca
1959													
Nov.	17	18.7	1.3	14.8	15.2	1.2	12.2	21.6	1.2	17.7	22.0	1.3	17.2
	30	17.4	1.2	14.2	15.6	1.2	13.4	24.2	1.2	19.8	21.7	1.3	17.0
Dec.	16	21.1	1.2	17.3	19.6	1.2	16.3	[b] 25.7	[a]		21.2	[a]	
	28	17.0	1.2	14.0	18.6	1.2	16.0	[b]	[b]		20.1	1.2	17.2
1960													
Jan.	11	19.2	1.3	15.0	12.5	1.2	10.6	30.0	1.3	23.8	24.0	1.1	21.8
	25	19.8	1.3	15.3	10.2	1.2	8.6	24.0	1.2	19.5	24.7	1.2	20.6
Feb.	9	17.8	1.2	15.4	13.6	1.2	11.3	24.0	1.2	19.8	25.9	1.2	21.9
	22	15.3	1.2	12.8	7.9	1.1	7.1	25.5	1.3	19.7	21.4	1.2	18.0
Mar.	7	16.5	1.1	15.2	9.2	1.2	7.8	27.0	1.1	24.5	22.5	1.2	18.3
	21	12.1	1.3	9.0	7.5	1.3	6.0	23.0	1.3	18.3	20.2	1.4	14.8
Apr.	4	8.5	1.2	7.1	8.3	1.2	7.2	27.6	1.2	22.9	21.5	1.3	16.9
	18	16.9	1.2	13.9	6.4	1.1	5.7	22.6	1.2	19.5	19.8	1.2	16.5
May	2	17.4	1.3	12.9	9.5	1.1	8.5	21.5	1.1	18.8	19.8	1.2	16.0
	16	10.6	1.2	9.9	7.4	1.1	6.8	45.0	1.2	39.0	25.3	1.2	20.8
	31	15.1	1.2	12.3	15.2	1.1	13.4	25.0	1.1	22.7	22.7	1.2	18.6
June	13	11.8	1.2	9.5	14.3	1.1	12.9	23.6	1.1	20.7	17.0	1.3	13.2
	27	17.1	1.0	16.5	7.5	1.2	6.3	24.1	1.2	20.3	22.0	1.1	19.8
Average		16.0	1.21		11.7	1.17		25.9	1.19		21.9	1.23	

[a] Analyses not made; sample lost during analysis.
[b] No sample collected.

Cattle Feeds

The data obtained from the analyses of the various feed samples are presented in tables 4, 5, and 6. These data are given in terms of micromicrocuries of strontium-90 per kilogram dry weight, grams of calcium per kilogram dry weight, and micromicrocuries of strontium-90 per gram of calcium. Instances where data do not appear for a specific sampling date indicate that the specific type of feed was not being fed.

In order to make some estimate of the total feed intake, the farm operator was questioned during each sampling period as to the daily regimen of the herd. These data are shown in table 6. The data are given in pounds per day per cow for each type of feed. Since the laboratory data are in a dry-weight basis, the data in table 7 are given both on a wet-weight and a dry-weight equivalent. These data are only the basis for an estimate of the relative importance of the various feeds in terms of strontium-90 intake. Comparison of

table 7 with the average levels of strontium-90 in the various feeds as shown in tables 4, 5, and 6 indicates that approximately 70 to 90 percent of the total strontium-90 intake came from the hay. This comparison indicates also that less than 5 percent of the strontium-90 and approximately 30 percent of the available calcium show up in the milk.

Table 4.--Concentrations of Strontium-90 and Calcium Found in Hay Feed

Date sampled		Farm 2(L)			Farm 10(L)			Farm 12(H)			Farm 14(H)		
		μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca
1959													
Nov.	17	1090	5.6	180	410	3.5	120	480	2.8	170	1610	7.9	205
	17	180	7.0	[a] 25									
	30	740	4.9	150	660	3.7	180	750	4.6	160	960	3.2	300
Dec.	16	820	5.6	150	390	7.9	50	420	6.3	68	750	4.3	180
	28	1170	4.1	270	310	2.8	106	420	3.9	107	1350	4.2	320
1960													
Jan.	11	450	6.8	66	390	3.5	[b] 110	1360	5.3	390	1020	4.7	220
	25	850	4.0	210	190	10.4	[b] 19	1580	6.8	230	1010	4.3	240
Feb.	9	670	6.5	110	390	9.3	[b] 42	790	4.2	190	800	4.0	200
	22	1120	5.4	210	670	12.6	[b] 54	1190	3.8	310	540	3.4	160
Mar.	7	1020	3.4	[c] 300	650	11.1	[b] 58	960	3.0	320	1260	3.4	370
	21	1100	4.2	[c] 250	560	23.3	[b] 24	1100	2.8	390	760	3.6	210
Apr.	4	250	6.2	[c] 40	530	13.1	[b] 40	1300	3.0	440	650	3.3	200
	18	830	8.1	[c] 100	560	11.8	[b] 49	820	4.2	200	1260	3.0	420
May	2	520	4.2	[c] 120	490	15.4	[b] 31	840	3.1	270	1300	6.7	190
	16	760	3.2	[c] 240	440	14.8	[b] 30	1100	3.0	360	900	3.9	230
	31												
June	13[d]	800	4.3	190	580	3.9	150	1510	4.8	310	360	2.4	150
	27[d]	1140	3.2	360	760	3.5	220	1400	3.5	400	1200	3.6	330
Average		795	5.1		499	9.4		1001	4.1		983	4.1	

Note: All samples are native grasses unless otherwise noted.
[a] Native alfalfa (farmer feeding some of each).
[b] Imported alfalfa.
[c] Half native grasses and half native alfalfa.
[d] Pasture grasses collected in pastures.

For comparative purposes with soil data, several feed samples were analyzed for total strontium content. The data obtained are shown in table 8. The figures in parentheses in this table are the levels of strontium-90 for the same samples in micromicrocuries per kilogram.

Water

The results of the analysis of drinking water are shown in table 9. The water, in all cases, was obtained from a ground water supply. No surface waters were available for cattle consumption during the study. This table includes the

Table 5.--Concentrations of Strontium-90 and Calcium in Corn Silage Feed

Date sampled	Farm 2(L)			Farm 10(L)			Farm 12(H)			Farm 14(H)		
	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca
1959												
Nov. 17	140	2.0	69	300	4.0	75	340	5.6	61	200	3.1	66
30	240	4.8	50	310	3.6	85	310	6.4	49	170	3.6	49
Dec. 16	300	6.4	46	300	5.2	59	350	7.9	45	170	5.5	32
28	290	4.6	64	270	3.8	72	370	5.4	68	208	3.1	67
1960												
Jan. 11	160	2.2	69	230	3.6	63	380	4.5	91	160	2.6	62
25	180	4.6	38	185	4.9	38	230	6.5	35	110	4.6	23
Feb. 9	450	5.2	88	310	4.2	72	270	3.9	69	250	5.2	47
22	240	3.6	66	260	3.6	74	310	4.8	65	270	5.0	54
Mar. 7	300	3.1	96	230	3.2	72	290	3.9	76	360	5.0	71
21	310	4.9	63	380	4.1	92	280	4.0	71	400	6.1	67
Apr. 4	290	3.4	86	330	4.2	78	360	4.9	73	400	6.0	67
18	300	3.4	88	360	4.9	74	430	4.7	92	310	3.6	87
May 2	200	3.2	60	280	4.4	64				340	4.6	74
16	280	2.7	100									
31	240	2.4	100									
June 13												
27												
Average	261	3.8		288	4.1		327	5.2		258	4.8	

Table 6.--Concentrations of Strontium-90 and Calcium Found in Grain Feed

Date sampled	Farm 2(L)			Farm 10(L)			Farm 12(H)			Farm 14(H)		
	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca	μμc Sr^{90}/kg (dry weight)	gm Ca/kg (dry weight)	μμc Sr^{90}/gm Ca
1959												
Nov. 17	106	4.2	25	73	5.6	13.1	12.5	.23	55	84	7.3	11.5
30	61	3.4	18.2	42	6.9	6.1	11.6	.18	66	39	5.0	7.8
Dec. 16	59	5.8	10.1	36	5.1	7.2	25	6.2	4.1	47	5.6	8.4
28	46	2.3	20	72	3.7	19.4	37	10.2	3.6	48	2.9	19.4
1960												
Jan. 11	35	1.6	22	72	6.6	11	31	1.4	22	57	2.7	21
25	32	5.1	6.3	42	4.9	8.5	40	7.8	5.1	47	4.0	11.9
Feb. 9	53	2.8	18.8	35	2.5	14.3	9.1	.17	55	6.8	2.1	32
22	66	3.8	17.2	38	1.9	19.4	130	1.0	120	41	3.1	13.4
Mar. 7	78	3.6	22	60	1.2	50	25	1.5	17	96	2.0	48
21	140	6.0	22	82	1.2	68	19	.8	23	120	3.6	33
Apr. 4	62	2.9	21	43	1.0	43	50	1.5	34	78	3.4	22
18	55	2.8	20	43	3.8	11.3	55	1.6	34	110	3.2	34
May 2	54	3.4	16	27	.85	32	21	.8	27	46	3.0	15
16	71	3.1	23	31	1.0	30	61	.7	88	71	3.1	23
31	64	2.6	25	23	1.1	22						
June 13	45	2.4	18	21	1.1	19				79	4.4	18
27	57	1.2	45	35	1.0	35				65	.8	84
Average	63.8	3.4		45.6	2.9		37.7	2.4		64.7	3.5	

Table 7.--Daily Feed Intake

Farm	Pounds fed per day per cow					
	Wet-weight basis			Dry-weight basis		
	Hay	Silage	Grain	Hay	Silage	Grain
2(L)	16	40	8	14	12	6
10(L)	25	22	10	23	7	7
12(H)	25	30	7	23	9	5
14(H)	15	40	10	14	12	7

Table 8.--Total Strontium in Selected Feed Samples

Sample		Total strontium (milligram per kilogram)			
Date	Type	Farm 2(L)	Farm 10(L)	Farm 12(H)	Farm 14(H)
Dec. 28, 1959	Grain		8.9 (72)	17.1 (37)	
	Silage		21 (270)	41 (420)	
	Hay		15 (310)	29 (420)	
	Silage	8.0 (450)	6.0 (310)	12.0 (270)	19.0 (250)
Feb. 9, 1960	Hay	31 (670)	13 (390)	13 (790)	29 (800)

date of sampling and the water analysis as gross beta activity in micromicrocuries per liter. An average for the entire sampling period is also included.

Table 9.--Gross Beta Activity Levels in Water Available to Cattle

Date sampled		Gross beta activity (μμc/liter)			
		Farm 2(L)	Farm 10(L)	Farm 12(H)	Farm 14(H)
1959					
November	17	*	*	6.5	6.5
	30	*	10.0	5.0	8.0
December	16	9.0	16.0	8.0	14.0
	28	*	*	*	1.0
1960					
January	11	*	*	*	4.0
	25	4.0	11.0	6.0	2.0
February	22	14.0	2.0	1.0	4.0
March	7	*	8.0	7.0	11.0
	21	*	*	*	18.0
April	4	4.0	15.0	10.0	18.0
	18	14.0	*	11.0	5.0
May	2	11.0	*	7.0	6.0
	16	15.0	10.0	19.0	9.0
	31	10.0	19.0	*	2.0
June	13	*	0.5	4.0	*
	27	*	*	*	11.0
Average		5.1	5.7	5.0	7.5

* Indistinguishable from background.

The data obtained from analysis of precipitation samples are presented in table 10. The data are given in terms

of liters of rain collected over a 0.4 square meter area, micromicrocuries of strontium-90 per liter, and total micromicrocuries of strontium-90 deposited during each sampling period. The total precipitation in inches of rain and the calculated total deposition of strontium-90 in terms of micromicrocuries per square meter for the entire period of collection are included in table 10. Where data are not shown, precipitation did not occur.

Table 10.--Concentrations of Strontium-90 in Precipitation

Sampling period (1960)	Farm 2(L)			Farm 10(L)			Farm 12(H)			Farm 14(H)		
	Volume (liter)	Sr^{90} (μμc/liter)	Sr^{90} deposited (μμc/0.4 m^2)	Volume (liter)	Sr^{90} (μμc/liter)	Sr^{90} deposited (μμc/0.4 m^2)	Volume (liter)	Sr^{90} (μμc/liter)	Sr^{90} deposited (μμc/0.4 m^2)	Volume (liter)	Sr^{90} (μμc/liter)	Sr^{90} deposited (μμc/0.4 m^2)
Feb. 29-Apr. 4	6.2	3.9	24.2	5.7	7.6	43.3	5.6	7.5	42.0	7.0	9.5	66.5
Apr. 4-11												
Apr. 11-18	11.0	2.3	25.3	8.5	2.4	20.4	7.2	2.3	16.6	8.8	2.8	24.6
Apr. 18-25	13.9	5.0	69.5	9.1	5.1	46.4	13.3	6.2	82.5	9.0	7.0	63.0
Apr. 18-May 2	5.2	2.7	14.0	1.7	2.0	3.4	5.0	2.5	12.5	2.7	2.8	7.6
May 2-9	13.1	4.0	52.4	8.3	5.9	49.0	9.7	4.0	38.8	10.2	6.8	69.4
May 9-16												
May 16-23	10.9	3.7	40.3	14.2	3.2	45.5	12.2	3.9	47.6	12.9	3.5	45.1
May 23-31	10.6	2.4	25.4	10.9	4.2	45.8	9.6	3.4	32.6	10.4	3.7	38.5
May 31-June 13	4.3	8.0	34.4	4.4	8.6	37.8	4.2	4.5	18.9	4.8	6.1	29.3
June 13-20	7.4	4.6	34.0				2.6	8.7	22.6	2.7	7.2	19.4
June 20-27	19.4	5.5	106.7	24.7	5.2	128.4	25.3	5.4	136.6	25.2	4.7	118.4
Total	102.0		426.2	87.5		420.0	94.7		450.7	93.7		481.8
Precipitation (inches)		10.1			8.6			9.3			9.2	
Sr^{90} deposited (μμc/m^2)		1065			1050			1125			1200	

Plant Species

Thirty-nine species of plants were identified in the pasture and marsh areas of the 4 farms (see Appendix VII, p. 71. Some measure of the abundance of a species is indicated by the number of X's. The most abundant species were bluegrass, timothy, panic grass, and white clover, with sedges common in the slough and marsh areas.

Vegetation samples were only partially representative of the soil sampling sites. Because insufficiency of

pasture growth made sampling impossible in some cases, the following compromises were made in choice of sites:

1. if an ungrazed area (fence row) was found within 200-250 feet of the soil sampling location, then it, rather than the pasture, was sampled for grass;
2. where areas of increased growth associated with cattle droppings were found, these were sampled; and
3. where areas of increased growth or fence rows could not be found, an unmeasured sample of the closely cropped pasture grass was taken.

DISCUSSION

The data obtained in this study, although in some cases quite approximate, do supply enough general information to serve the basic purpose of the project. The data regarding the concentrations of strontium-90 in milk, as shown in table 3, indicate significant farm-to-farm differences persisting throughout the sampling period. Thus, this discussion will attempt to relate the differences in the milk to the differences that exist in the intake of the cattle.

Statistical analysis showed essentially no correlation between the concentrations of strontium-90 and calcium in the individual feed samples and the concentrations found in the milk sampled at the same time. This was not surprising since both feed and milk were grab samples and only very approximate feeding data were available. Such data indicated, however, that less than 5 percent of the strontium-90 available in the feed showed up in the milk.

It is important to note that the drinking water may be disregarded in terms of strontium-90 intake as far as these data are concerned. The low levels of gross beta activity found (see table 9) are quite insignificant in comparison to the concentrations of strontium-90 found in the feeds.

From tables 4, 5, and 6 it can be seen that the concentrations of strontium-90 found in the native grass hay samples were, in general, much higher than the concentrations found in either grain or silage samples. The average strontium-90 concentrations in the hay were approximately two to four times the concentrations in silage and approxi-

mately one order of magnitude higher than the concentrations found in grain. Thus it would be suspected that the hay contributed the greatest proportion of the strontium-90 to the cattle's intake. This was further substantiated by the daily intake data in table 7. Using these data, together with the average levels of strontium-90 in the feeds, it was found that approximately 70 to 90 percent of the strontium-90 intake came from the hay.

Since it is reasonably apparent that the hay should contribute the greatest proportion of the strontium-90 to the cattle's diet, one would expect to find a correlation between the farm-to-farm differences in milk and the farm-to-farm differences in hay. This, in fact, appeared to be the case. That such a relationship existed can be seen by comparing the average concentrations of strontium-90 in milk (micromicrocuries per liter) to the average concentrations in hay (micromicrocuries per kilogram) for the 4 farms (tables 3 and 4). It is apparent that the over-all average concentrations in hay were reasonably indicative of the over-all average concentrations in milk. Statistical comparison of these mean data yielded a linear correlation coefficient of 0.93.

To further analyze the relationship between the concentration in milk and in hay, running averages were obtained by averaging the concentration in a given sample with the sample immediately preceding it and the sample immediately following it. Running averages of the hay (in barn feeding only) and milk data for farms 10(L) and 12(H) were calculated and compared on an individual farm basis. Although the individual samples had shown no apparent correlation, the running averages produced better results. Correlation coefficients of 0.77 for farm 10(L) and 0.71 for farm 12(H) were obtained by relating the ratio of strontium-90 to calcium in the hay to that of strontium-90 to calcium in the milk when these running averages were used. These data are plotted in figure 3.

Too much importance should not be placed on the correlation coefficient alone, since the number of samples compared was quite small, and thus the error might be large.

Considering the higher concentrations in the hay, the high estimated intake of strontium-90 from the hay, and the correlation between the concentrations of strontium-90 in the hay and in the milk, it seems reasonable that farm-to-farm differences in the milk produced can be attributed primarily to like differences in the hay fed.

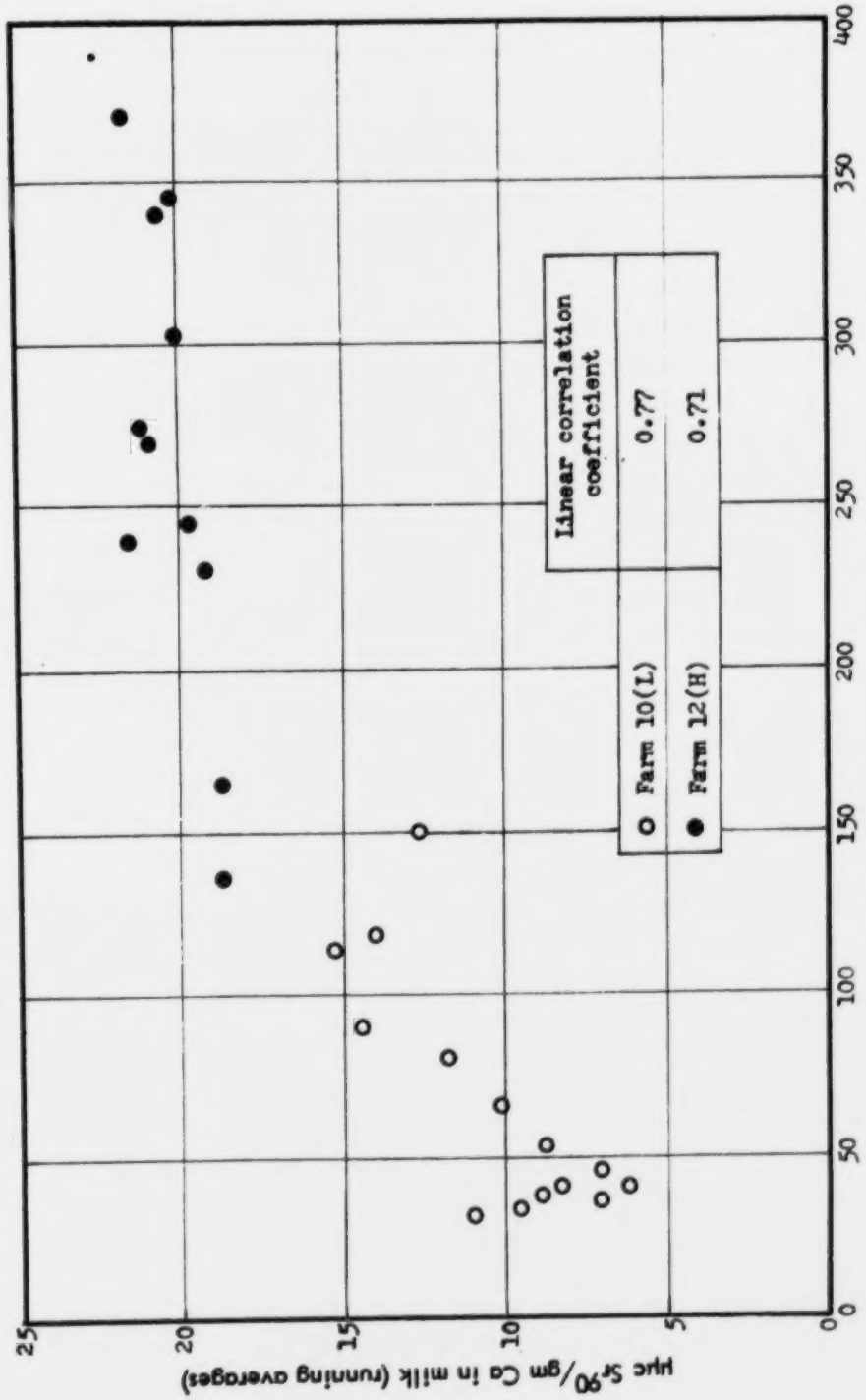

Figure 3.--Correlation of Strontium-90 to Calcium Ratios in Milk and Hay

During part of this study, both low farms fed quantities of alfalfa hay while the high farms were feeding only native grasses. Part of the differences in the milk concentrations may have been caused simply by this difference in the type of hay being fed. Table 4 indicates that the alfalfa being fed on farm 10(L) showed much higher calcium content and thus much lower strontium-90 to calcium ratios than the native grasses being fed previously. One would expect this decrease in the feed strontium-90 to calcium ratio to be reflected in the concentrations of strontium-90 found in the milk.

Although a part of the differences in the milk may be attributed to the feeding of a high calcium alfalfa, this does not seem to be the complete answer. If only the native grass samples from table 4 are compared, one still finds sizeable differences from farm to farm. Table 11 shows these average concentrations in terms of micromicrocuries of strontium-90 per kilogram and micromicrocuries of strontium-90 per gram of calcium.

Table 11.--Strontium-90 Content of Native Grasses (Averages)

Feed	Farm 2(L)		Farm 10(L)		Farm 12(H)		Farm 14(H)	
	μμc/kg	μμc/gm Ca	μμc/kg	μμc/gm Ca	μμc/kg	μμc/gm Ca	μμc/kg	μμc/gm Ca
[a]	885	190	500	134	1000	270	983	245
[b]	864	168	432	113	936	257	1010	246

[a] Native grasses including pasture samples taken June 13, 1960, and June 27, 1960.
[b] Native grass hay only.

It does not appear that the farm-to-farm differences in the native grass hay could be explained on the basis of differences in fallout or rainout of strontium-90. Precipitation samples collected and analyzed over the period of sampling show only small differences in the total deposition of strontium-90 (table 10). The hay had, of course, been grown the previous year, for which, unfortunately, precipitation data are not available. However, pasture grass samples obtained for this study showed farm-to-farm differences in the concentrations of strontium-90 in native grasses of approximately the same magnitude as those observed in the hay grown the previous year. During this period there were no great differences in strontium-90 deposition. Table 4 contains several pasture grass samples obtained during this study. Several additional samples were obtained after the study had ended. Table 12 presents a comparison of the strontium-90 concentrations in pasture grass samples for farms 10(L) and 12(H).

Table 12.--Concentrations of Strontium-90 in Pasture Grasses (μμc/kg)

Date sampled	Farm 10(L)	Farm 12(H)
1960		
June 13	580	1510
27	760	1400
July 25	850	1600
August 9	860	*
23	*	1050

* Samples not collected.

Thus, differences in the concentrations of strontium-90 in the native grasses existed during a period when there were no significant differences in strontium-90 deposition.

CONCLUSIONS

1. Farm-to-farm differences of approximately a factor of 2 in concentrations of strontium-90 in milk persisted throughout the study.

2. Seventy to 90 percent of the total strontium-90 intake of the cattle apparently came from the native grass hay fed.

3. Drinking water was insignificant in terms of strontium-90 intake.

4. Farm-to-farm differences in the concentrations of strontium-90 in the milk appeared to be the result of like differences in the hay fed.

5. The total deposition of strontium-90 by rain during the study did not differ to any great extent between the farms.

Part II

RESULTS

Soils

Soils identification. Some general information regarding the soil samples obtained in this study is presented in

table 13. The soil sample designations are used throughout this report to permit ready identification of the individual samples for cross reference. A detailed description of the soil series is given in Appendix III, p. 56.

Table 13.--Identification of Soils

Farm	Soil sample designation*	Location description	Depth (inches)	Soil series
10(L)	18-U-CF	Upland (cornfield)		
	19-U-S	Upland surface	0-2	Brainerd
	20-U-SS	Upland subsurface	2-8	Brainerd
	21-M-S	Marsh surface	0-2	Barrows
	22-M-SS	Marsh subsurface	2-8	Barrows
2(L)	23-M-S	Marsh surface	0-2	Barrows
	24-M-SS	Marsh subsurface	2-8	Barrows
	25-L-S	Lowland surface	0-2	Nokay
	26-L-SS	Lowland subsurface	2-8	Nokay
	27-U-S	Upland surface	0-2	Brainerd
	28-U-SS	Upland subsurface	2-8	Brainerd
12(H)	29-U-S	Upland surface	0-2	Brainerd
	30-U-SS	Upland subsurface	2-8	Brainerd
	31-L-S	Lowland surface	0-2	Nokay
	32-L-SS	Lowland subsurface	2-8	Nokay
	33-M-S	Marsh surface	0-2	Barrows
	34-M-SS	Marsh subsurface	2-8	Barrows
14(H)	35-M-S	Marsh surface	0-2	Barrows
	36-M-SS	Marsh subsurface	2-8	Barrows
	37-L-S	Lowland surface	0-2	Nokay
	38-L-SS	Lowland subsurface	2-8	Nokay

*U - Upland
L - Lowland
M - Marsh
S - Surface soil
SS - Subsurface

Soils characteristics. Commonly determined chemical characteristics in the routine soil testing procedure are pH, organic matter, available phosphorous, and available potassium. ("Available" as used here is a relative term and indicates the probable availability of a nutrient to plants compared with other soils.) These data for the various soil samples are presented in table 14.

Data showed that these soils were acidic in nature down to the 8-inch level sampled. A pH of 6.0 does not represent a condition of acidity in the soil that would be particularly detrimental to plant growth. A pH of 4.9, on the other hand, represents quite an acid condition. In general, these soils were moderately high in organic matter. Sample 18-U-CF was low in organic matter, but this surface sample was from a cultivated field while the other soil samples were from uncultivated fields and pastures.

Table 14.--Characteristics of Soils

Farm	Soil number	pH	% organic matter	P/A[a] (lbs.)	K/A[b] (lbs.)
10(L)	18-U-CF	6.1	2.2	96	100
	19-U-S	6.2	5.3	30	250
	20-U-SS	6.0	4.4	19	70
	21-M-S	5.4	8.5+	25	240
	22-M-SS	5.3	4.9	21	50
2(L)	23-M-S	4.9	8.5+	50	260
	24-M-SS	5.2	6.1	100	60
	25-L-S	5.8	4.9	36	410
	26-L-SS	6.2	4.1	30	110
	27-U-S	6.0	8.5+	37	330
	28-U-SS	5.8	2.2	30	80
12(H)	29-U-S	6.1	4.7	39	340
	30-U-SS	6.0	3.1	34	120
	31-L-S	5.7	8.5+	35	260
	32-L-SS	5.7	3.6	49	70
	33-M-S	5.0	8.5+	110	300
	34-M-SS	5.2	8.5+	200+	160
14(H)	35-M-S	5.4	8.5+	37	260
	36-M-SS	5.4	4.0	80	50
	37-L-S	5.5	8.5+	40	340
	38-L-SS	6.0	2.3	26	60

[a] P/A - available phosphorous.
[b] K/A - available potassium.

Values for available phosphorous (P/A) indicated relatively high amounts of this element. Available potassium (K/A) values of less than 100 represent a deficiency of this nutrient.

Exchangeable calcium and magnesium and total exchange capacity of soils. Exchangeable calcium (Ex. Ca) and magnesium (Ex. Mg) and the total exchange capacity of the soils in terms of parts per million (ppm) and milliequivalents per 100 grams (me/100 gm) are given in table 15. (The major source of calcium and magnesium for plants on acid soils is that which is adsorbed on the exchange complex.) Total exchange capacity represents the total capacity of the adsorbing material of the soil to retain exchangeable cations. In these soils, calcium and magnesium made up less than a third of the exchangeable cations. Low amounts of exchangeable calcium were also indicated by the low pH's of these soils (table 15).

Except for one or two instances, amounts of exchangeable calcium were of about the same orders of magnitude as the calcium extracted by 6N hydrochloric acid.

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Table 15.--Exchangeable Calcium and Magnesium and Total Exchange Capacity in Soils

Farm	Soil number	Ex. Ca		Ex. Mg		Total ex. cap. (me/100 gm)
		ppm	me/100 gm	ppm	me/100 gm	
10(L)	18-U-CF	562	2.8	ND*	ND	12
	19-U-S	1095	5.5	ND	ND	18
	20-U-SS	545	2.7	ND	ND	12
	21-M-S	1050	5.2	ND	ND	28
	22-M-SS	375	1.9	54	0.4	14
2(L)	23-M-S					ND
	24-M-SS	387	1.9	60	0.5	18
	25-L-S	875	4.4	108	0.9	27
	26-L-SS	900	4.5	117	1.0	15
	27-U-S	1172	5.8	164	1.3	25
	28-U-SS	240	1.2	46	0.4	11
12(H)	29-U-S	587	2.9	74	0.6	14
	30-U-SS	435	2.2	44	0.4	12
	31-L-S	1630	8.1	156	1.3	26
	32-L-SS	432	2.2	41	0.3	11
	33-M-S	1125	5.6	182	1.5	ND
	34-M-SS	885	4.4	149	1.2	ND
14(H)	35-M-S	1225	6.1	157	1.3	35
	36-M-SS	335	1.7	79	0.7	17
	37-L-S	1412	7.0	165	1.4	ND
	38-L-SS	612	3.1	71	0.6	ND

* ND - not determined.

Total calcium and stable strontium. Total calcium and strontium in terms of parts per million and millimoles per kilogram of oven dry soil are given in table 16.

Statistical comparisons of the data show that strontium concentrations did not vary significantly between surface and subsurface samples (table 17). Calcium concentrations, however, were lower at the 99 percent confidence level in subsurface soils than in surface soils.

Statistical comparisons were also made of strontium and calcium concentrations in soils from farms supplying high strontium-90 concentrations in milk with those of low concentrations (table 18). There were significantly lower strontium concentrations in soils from low strontium-90 suppliers than from high suppliers, in both surface and subsurface soils though this difference was more pronounced in surface soils. Calcium concentrations did not vary significantly in either surface or subsurface soils between the 2 groups of farms.

The statistical correlation of calcium with strontium is shown in figure 4. A correlation coefficient of $r = 0.531$ was found which was different from zero at greater than the

Table 16.--Calcium and Strontium Values Observed in Soils

Farm	Soil number	ppm Ca	ppm Sr	m mole Ca/kg	m mole Sr/kg
10(L)	18-U-CF	576	3.7	14.4	0.04
	19-U-S	1324	12.5	33.0	0.14
	20-U-SS	838	11.4	20.9	0.13
	21-M-S	983	7.1	24.5	0.08
	22-M-SS	347	10.2	8.6	0.12
2(L)	23-M-S	677	8.6	16.9	0.10
	24-M-SS	810	7.5	20.2	0.09
	25-L-S	956	7.4	23.8	0.08
	26-L-SS	985	7.6	24.6	0.09
	27-U-S	2100	10.0	52.4	0.11
	28-U-SS	311	5.4	7.7	0.06
12(H)	29-U-S	656	10.2	16.4	0.12
	30-U-SS	452	9.8	11.3	0.11
	31-L-S	1253	14.8	31.2	0.17
	32-L-SS	441	11.1	11.0	0.13
	33-M-S	1220	9.6	30.4	0.11
	34-M-SS	840	15.7	20.9	0.18
14(H)	35-M-S	1105	10.3	27.6	0.12
	36-M-SS	315	10.6	7.8	0.12
	37-L-S	1960	30.6	48.9	0.35
	38-L-SS	642	5.1	16.0	0.06

Table 17.--Comparison of Stable Strontium and Calcium Content of Surface and Subsurface Soils

Contrast	Surface (m moles/kg)	Subsurface (m moles/kg)	Significance probability
Strontium	0.14	0.11	NS*
Calcium	30.5	14.9	99.0%

* NS - non-significant

Table 18.--Comparison of Strontium and Calcium Concentrations in Soils from Farms High and Low in Strontium-90

Element	Soil location	High Sr^{90} farms	Low Sr^{90} farms	Significance probability
		m moles/kg		
Strontium	Surface	0.17	0.10	95%
Strontium	Subsurface	0.12	0.10	73%
Calcium	Surface	30.9	30.1	NS*
Calcium	Subsurface	13.4	16.4	NS

* NS - non-significant

95 percent confidence level. The value of r^2 was 0.282, indicating that 28.2 percent of the variation in strontium concentrations was associated with variation in calcium concentrations.

Cesium-137 content of soils. The amounts of strontium-90 as determined by electrodialysis extraction of soils

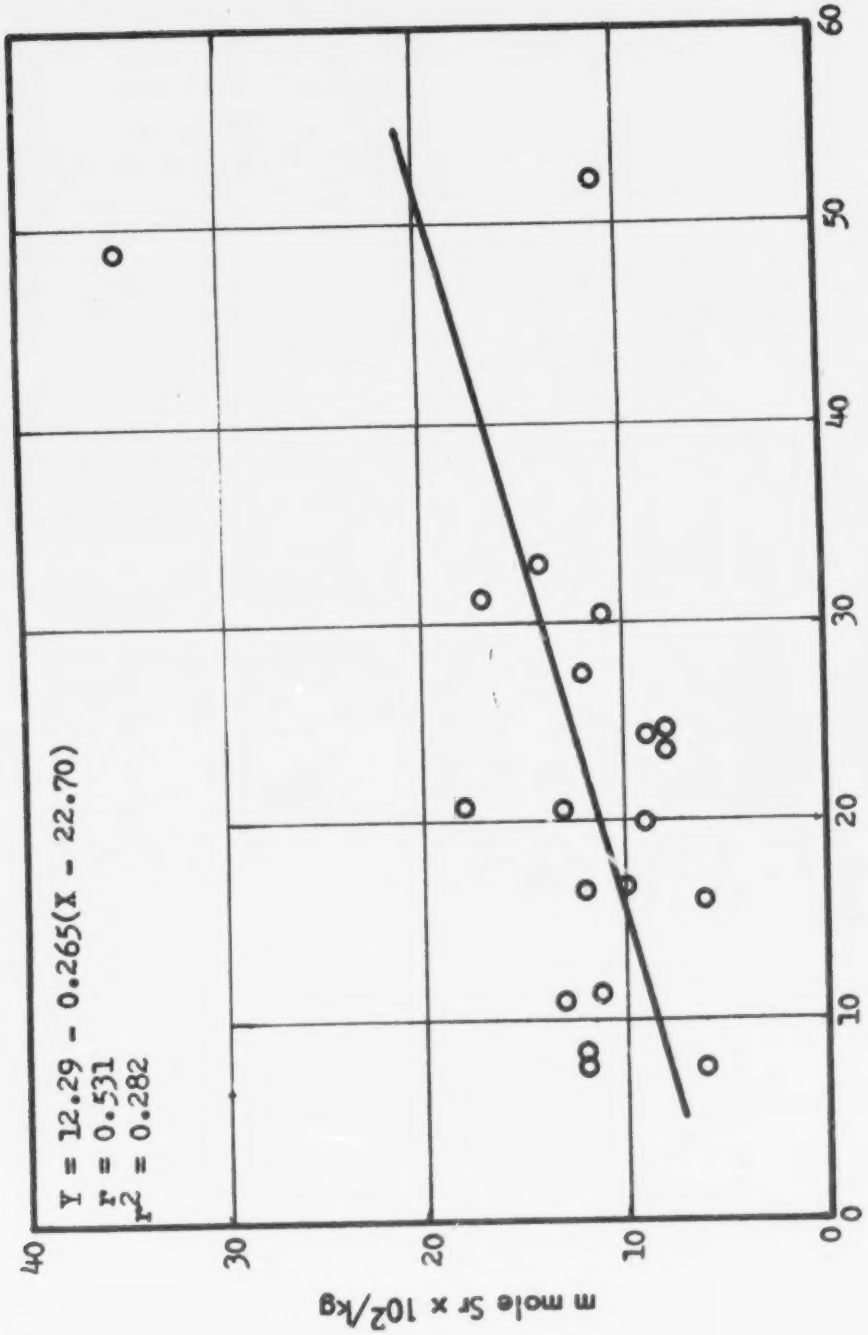

Figure 4.--Correlation of Strontium with Calcium in Soils

appeared to be unreliable. Dr. P. F. Gustafson, Argonne National Laboratory, Lemont, Illinois, has suggested that a good estimate of the strontium-90 content of soils could be obtained by determining cesium-137, values of which are about twice those for strontium-90. Because supplies of soils collected in the fall of 1959 were depleted, cesium-137 was run on samples collected in the spring of 1960 from the identical sites. These analyses, from which strontium-90 can be approximated, were run at the Argonne National Laboratory. The cesium-137 values in micromicrocuries per kilogram and millicuries per square mile are given in table 19.

Table 19.--Cesium-137 Content of Soils

Farm	Soil number*	Cs-137	
		μμc/kg	mc/mi^2
10(L)	19-U-S	336	44.4
	21-M-S	1736	93.4
2(L)	23-M-S	1642	100.6
	27-U-S	620	53.6
12(H)	29-U-S	354	52.8
	31-L-S	624	57.4
14(H)	35-M-S	1446	126.6
	37-L-S	400	30.0

* Though soil numbers listed more properly identify fall collected samples, in this table they are used to designate samples taken in the spring. This is justified on the basis that sampling sites for fall and spring collected samples were identical.

Amounts of cesium-137 in marsh areas (Barrows soil) were several times greater than in soils which lie somewhat higher topographically.

Strontium-90 content of soils. The strontium-90 content of some of the soils from the farms is given in table 20. The term "soil" as used in this section refers to the surface soil minus the undecomposed organic matter, which is called the "root mat" in the succeeding section. Strontium-90 was determined in 3 ways: viz., in soil extracts obtained by electrodialysis and separated on ion exchange columns; in soil extracts using 6N hydrochloric acid; and from cesium-137 values (a cesium-137 to strontium-90 ratio of 2:1 has been found in a number of soil samples to approximate fallout distribution).

Electrodialysis and 6N hydrochloric acid extracts were made on soils collected in the fall of 1959. Cesium-137 determinations were made on soil samples from exactly the same sites collected in the spring of 1960. Since values reported

were on soils from which root mats were removed, they were comparable in this respect also. Electrodialysis-ion exchange analyses are not included in table 20 because of the uncertainty of the values obtained. A discussion of the electrodialysis method and analytical results is given in Appendix IV, p. 57.

Comparable data for strontium-90 content determined by 2 independent methods were available for only 2 sites, 27 and 29. While these data were meager, it was interesting to note the close agreement between the results of the 2 methods of analysis (table 20).

Table 20.--Strontium-90 Content of Soils

Farm	Soil number	Sr^{90} in HCl extracts[a]		Sr^{90} from Cs^{137}/Sr^{90} ratios[b]	
		μμc/kg	mc/mi^2	μμc/kg	mc/mi^2
10(L)	19-U-S			168	22.2
	21-M-S			868	46.7
2(L)	23-M-S			821	50.3
	25-L-S	150	22.2	-	-
	27-U-S	290	26.3	310	26.8
12(H)	29-U-S	160	24.7	177	26.4
	31-L-S			312	28,7
	33-M-S			-	-
14(H)	35-M-S			723	63.3
	37-L-S			200	15.0

[a] Determined in fall collected samples.
[b] Determined in spring collected samples from exactly the same sites as soils taken in the fall.

There were large differences in amounts of strontium-90 in these soils. However, there was no apparent relationship between amounts of strontium-90 in the soils and whether or not the milk from the farms was high or low in strontium-90.

There was an indication from strontium-90 values estimated from cesium-137 that the marsh areas were higher in strontium-90 than were higher topographic locations.

Vegetation

Strontium-90 and calcium content of root mats. The strontium-90 and calcium contents of root mats are given in table 21.

Calcium concentrations in the various root mats were not greatly different, except for sample 27. Reference to table 16 will show that sample 37 had a relatively high calcium

content in the soil also. The calcium content of low farms did not seem to be significantly different from high farms. Strontium-90 contents were variable, but no particular trend was apparent to distinguish between high and low farms. Also reported were strontium-90 contents in millicuries per square mile. Table 21 indicates that the root mat contained much less strontium-90 than did the soil from which it was removed.

Table 21.--Calcium and Strontium-90 Content of Fall Sampled Root Mats

Farm	Soil number	Ca (gm/kg)	Sr^{90}	
			μμc/kg	mc/mi^2
10(L)	19-U-S	3.1	1300	1.6
	21-M-S	3.7	2800	2.9
2(L)	23-M-S	3.6	2500	3.0
	25-L-S	2.1	660	0.9
	27-U-S	10.0	3000	6.4
12(H)	29-U-S	3.3	1200	1.7
	31-L-S	3.6	1850	1.9
	33-M-S	3.2	1300	0.7
14(H)	35-M-S	4.9	2000	2.5
	37-L-S	5.8	2700	3.4

Calcium, stable strontium, and strontium-90 in vegetation obtained from soil sampling sites. Stable (or natural) strontium was determined in the vegetation secured in the spring from some of the soil sampling sites (table 22). There was no significant difference in the amount of stable strontium between the farms on which high or low strontium-90 milk was found.

Calcium was significantly less in spring vegetation than in fall vegetation, although high and low farms did not differ. Strontium-90, on the other hand, was higher in spring vegetation. High and low farms did not differ significantly in strontium-90 content of vegetation. Amounts of strontium-90 in millicuries per square mile in the fall vegetation from some of the soil sampling sites are given in table 22. (In order to determine millicuries of strontium-90 per square mile of vegetation, it is necessary to have a measure of the weight of the grass per unit area. Sufficient grass was secured for analysis, but in some of the fall sampled sites and all of the spring sampled areas, vegetation was so sparse that a quantitative estimate of yield per unit area was not possible.) Strontium-90 in root mats can be regarded as an estimate of the strontium-90 in vegetation from the same site, since root mats separated from the soils included part of the closely cropped surface vegetation. Strontium-90 concentrations in root mats and vegetation were of the same

order of magnitude (tables 21 and 22).

Data on vegetation from high and low farms were insufficient for drawing conclusions. Strontium-90 content varied with topographic location, however. Strontium-90 was significantly higher in vegetation obtained from marsh sites than in that from other sites.

Table 22.--Stable Strontium, Calcium, and Strontium-90 in Vegetation Obtained from Soil Sampling Sites in November 1959 and May 1960

Farm	Soil number	Stable Sr	Calcium		Strontium-90		
		Spring 1960	Fall 1959	Spring 1960	Fall 1959		Spring 1960
		(mg/kg)	(gm/kg)	(gm/kg)	μμc/kg	mc/mi^2	(μμc/kg)
10(L)	19-U	16	5.2	3.6	1740		2000
	21-M	25	5.0	4.0	2560	1.7	3400
2(L)	23-M	18	4.0	3.3	1790	1.5	2200
	25-L	-	6.0	-	1330	0.3	-
	27-U	24	5.2	4.4	1930	1.6	2700
12(H)	29-U	23	3.7	3.8	1300	-	2000
	31-L	26	5.4	3.8	1380	0.3	2800
	33-M	-	5.2	-	1610	0.6	-
14(H)	35-M	25	5.8	4.8	3290	-	5200
	37-L	26	5.8	5.2	1520	-	2100

Soils and Vegetation

Strontium-90 to calcium ratios in soils, root mats, and vegetation. Table 23 provides the strontium-90 to calcium ratios in soils, root mats, and vegetation. Ratios for soils, root mats, and fall vegetation were higher on the average for the farms with low strontium-90 concentrations in the milk; for spring vegetation, just the reverse was true. The fall root mat had a higher strontium-90 to calcium ratio than the fall vegetation at the 98 percent level of significance. Ratios in spring vegetation were significantly higher than those in fall vegetation. Ratios in spring vegetation were, in general, several times higher than those of soils from the same site (table 23). Strontium-90 to calcium ratios for soils, root mats, and vegetation were significantly higher on the marsh sites than on other sites.

Stable strontium to calcium ratios in soils and vegetation. Stable strontium to calcium ratios are given in table 24. The ratios in soils ranged from 4.8 to 33.6 milligrams of strontium per gram of calcium and were considerably more variable than the ratios in vegetation taken from the same sites.

A statistical treatment of the data is given in table 25.

Table 23.--Strontium-90 to Calcium Ratios in Soils, Root Mats, and Vegetation

Farm	Soil number	Soils* (Spring 1960)	Root mats (Fall 1959)	Vegetation	
				Fall 1959	Spring 1960
		μμc Sr^{90}/mg Ca			
10(L)	19-U-S	127	430	330	550
	21-M-S	883	750	510	860
2(L)	23-M-S	1210	680	450	650
	25-L-S	-	310	210	-
	27-U-S	147	310	370	620
12(H)	29-U-S	270	370	360	520
	31-L1S	249	570	260	740
	33-M-S	-	420	320	-
14(H)	35-M-S	655	410	570	1080
	37-L-S	102	470	260	400

* Ratios calculated using Sr^{90} values from Cs^{137} determinations. These are spring samples, from the same sites as those taken in the fall.

Table 24.--Stable Strontium to Calcium Ratios in Soils and Vegetation

Farm	Soil number	Soil[a] (mg Sr/gm Ca)	Vegetation[b] (mg Sr/gm Ca)
10(L)	18-U-CF	6.4	-
	19-U-S	9.4	4.4
	20-U-SS	13.6	-
	21-M-S	7.2	6.3
	22-M-SS	29.4	-
2(L)	23-M-S	12.7	4.5
	24-M-SS	9.3	-
	25-L-S	7.7	-
	26-L-SS	7.7	-
	27-U-S	4.8	5.5
	28-U-SS	17.3	-
12(H)	29-U-S	15.0	6.1
	30-U-SS	21.6	-
	31-L-S	11.8	6.8
	32-L-SS	25.2	-
	33-M-S	7.8	-
	34-M-SS	18.7	-
14(H)	35-M-S	9.3	5.2
	36-M-SS	33.6	-
	37-L-S	16.0	5.0
	38-L-SS	8.0	-

[a] Samples collected in the fall, 1959.
[b] Samples collected in the spring, 1960.

Comparisons were made between ratios from surface and subsurface soils and between ratios from high and low farms. It was found that strontium to calcium ratios were significantly higher in subsurface soils than in surface soils. The higher subsurface soil ratios primarily reflect decreased calcium concentrations in the subsurface soils and were influenced

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only slightly by variation in strontium from surface to subsurface (table 25).

Table 25.--Statistical Comparison of Strontium to Calcium Ratios in Soils

Soil location	Farms high in Sr^{90}	Farms low in Sr^{90}	Significance probability	Average ratio	Significance probability
Surface	11.9	8.3	93%	10.1	98%
Subsurface	21.4	15.9	85%	18.1	

When strontium to calcium ratios were compared between high and low farms, it was found that the ratios were significantly higher on the high farms (table 25). In this case the higher ratios were a reflection of higher stable strontium concentrations in soils from farms producing milk high in strontium-90, since average calcium concentrations between the farms were not different (table 16). When surface soils were compared, differences between ratios obtained from high and low farms were significant at a relatively high confidence level. Differences in the subsurface soils were consistant with those in the surface but were significant at somewhat lower confidence levels.

Stable strontium to calcium ratios of the vegetation from soil sampling sites were fairly uniform, with the 4 samples from the high farms averaging slightly higher (5.8 mg/gm) than the 4 samples from the low farms (5.1 mg/gm)(table 24). With one exception (sample 27), strontium to calcium ratios in vegetation were less than those in soils.

The relationships between strontium to calcium ratios and strontium and calcium concentrations in soils are shown in figures 5 and 6, respectively. A significant correlation was found between the strontium to calcium ratio and strontium with a correlation coefficient of 0.88. The ratio increased with increasing strontium, but not at a rate which would be expected if the value of the ratio were a function of strontium levels only.

The strontium to calcium ratio decreased with an increase in calcium concentration in the soil (figure 6). This relationship can be described by an equation of the form: $(Sr/Ca) \times 10^3 = 16.5 - 0.707Ca + 0.0093Ca^2$. The form of the equation indicates that the value of the ratio was not completely a function of calcium levels.

Strontium-90 to stable strontium ratios in soils and

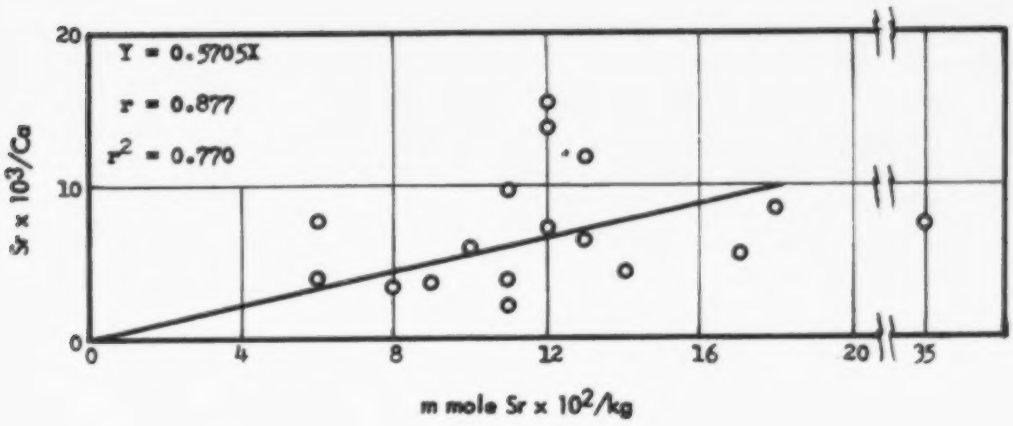

Figure 5.--Relationship Between Mole Ratios of Strontium and Calcium and Levels of Strontium in Soils

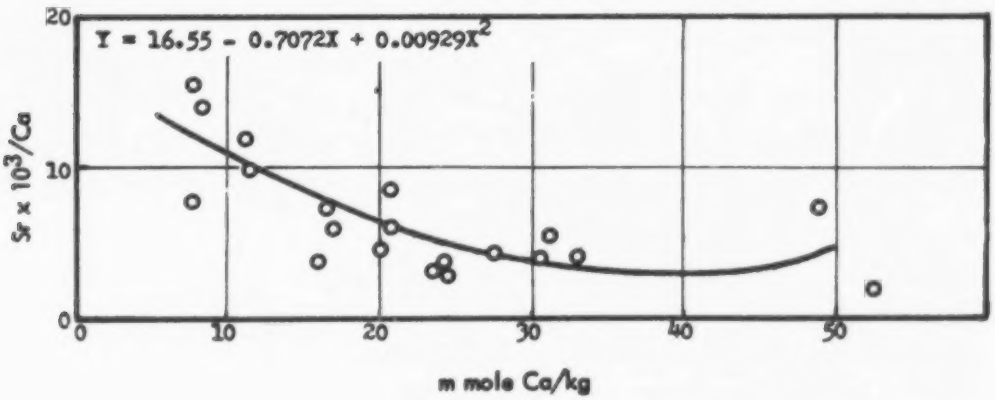

Figure 6.--Relationship Between Mole Ratios of Strontium and Calcium and Levels of Calcium in Soils

vegetation. Strontium-90 to stable strontium ratios of spring collected soils and vegetation are given in table 26. (Stable strontium was determined in fall collected soil samples only, and these values were used in calculating the ratios in table 26. The assumption was that stable strontium would not be greatly different in soils collected at different times from identical sites.) There was great variation in the strontium-90 to strontium ratios of soils, with no particular pattern for high or low farms.

Ratios for vegetation were more uniform, and were universally higher, sometimes several fold, than corresponding soil ratios. The magnitudes of the vegetation ratios did not correspond to those in soils from the same sites.

The ratios of strontium-90 to strontium were significantly higher in the soil and vegetation on the marsh sites than on the upland and lowland sites.

Table 26.--Strontium-90 to Stable Strontium Ratios of Soils and Vegetation, Spring 1960

Farm	Soil number	$\mu\mu c\ Sr^{90}/mg\ Sr$	
		Soil	Vegetation
10(L)	19-U-S	13.5	125
	21-M-S	122.6	136
2(L)	23-M-S	95.3	122
	25-L-S	-	-
	27-U-S	30.6	112
12(H)	29-U-S	18.0	86
	31-L-S	20.8	108
	33-M-S	-	-
14(H)	35-M-S	70.4	208
	37-L-S	6.4	81

Soil contrasts. Table 27 compares chemical data from the marsh sites with those from lowland and upland sites. No difference between the 2 soil groups was found in exchangeable calcium or magnesium, 6N hydrochloric acid extractable calcium, or stable strontium contents of surface soil, nor in total calcium or strontium content of root mat or vegetation. The strontium-90 contents, strontium-90 to calcium ratios, and strontium-90 to strontium ratios of the spring sampled soils were higher in the Barrows than in the Nokay and Brainerd soils. Although the strontium-90 content of the root mat was not measurably different, the strontium-90 to calcium ratio was higher at the marsh sites. The strontium-90 contents, strontium-90 to calcium ratios, and strontium-90 to strontium ratios were higher in vegetation taken from the Barrows soil than in that taken from the better drained soils.

The Barrows soil areas were the most highly contaminated parts of the landscape.

Table 27.--Some Contrasts Between Barrows Soil Sites (Marsh), Brainerd Soil Sites (Upland), and Nokay Soil Sites (Lowland)

Contrasts	Upland and lowland sites	Marsh sites	Significance probability
me exchangeable Ca/100 g surface soil	5.62	5.65	NS*
me exchangeable Mg/100 g surface soil	1.09	1.40	NS
ppm Ca in surface soil	1375	996	NS
ppm Sr in surface soil	14.2	8.9	NS
mg Sr/gm Ca in fall collected surface soil	10.8	9.2	NS
gm Ca/kg root mat	4.6	3.8	NS
gm Ca/kg fall sampled vegetation	5.2	5.0	NS
gm Ca/kg spring sampled vegetation	4.2	4.0	NS
mg Sr/kg spring sampled vegetation	23	23	NS
mg Sr/gm Ca in vegetation	5.6	5.3	NS
$\mu\mu c$ Sr^{90}/kg surface soil from Cs^{137} content	233	804	98%
mc Sr^{90}/mi^2 surface soil from Cs^{137} content	23.8	53.4	98%
$\mu\mu c$ Sr^{90}/gm Ca in spring sampled surface soil	179	916	98%
$\mu\mu c$ Sr^{90}/mg Sr in spring sampled surface soil	17.9	96.1	98%
$\mu\mu c$ Sr^{90}/kg root mat	1785	2150	NS
$\mu\mu c$ Sr^{90}/gm Ca in root mat	410	565	99%
$\mu\mu c$ Sr^{90}/kg fall sampled vegetation	1533	2312	97%
$\mu\mu c$ Sr^{90}/kg spring sampled vegetation	2320	3600	93%
$\mu\mu c$ Sr^{90}/gm Ca in fall sampled vegetation	298	462	97%
$\mu\mu c$ Sr^{90}/gm Ca in spring sampled vegetation	566	863	96%
$\mu\mu c$ Sr^{90}/mg Sr in spring sampled vegetation	102	155	96%

* NS - non-significant.

DISCUSSION

Data on soils and crops in this study were obtained in an attempt to explain the variation in the amount of strontium-90 in milk from the different farms. This survey represents an exploratory type of study, rather than an experimental one under which some controls can be set up.

Soil Characteristics

Some of the chemical properties of the soils were characterized by determination of pH, organic matter, and available phosphorous and potassium. The pH of the soils varied from moderately acid (6.1) to strongly acid (4.9). These data have major significance only to the extent that a low pH indicates the presence of few bases (Ca, Mg, K, and Na) and thus less competition is offered to the entry of strontium into the plant.

Organic matter concentrations were moderate to high. High organic matter indicates large capacity for adsorbing or holding cations such as strontium. Available phosphorous and potassium would seem to have little significance to this study. In summary, chemical characteristics were not greatly different in farms with high or low strontium-90 in milk, and thus no clue is offered to explain the variation in strontium-90 content.

Since calcium and magnesium would be expected to offer competition to strontium in the absorption of bases from the soil, exchangeable calcium and magnesium were determined as a measure of the availability of these nutrients to plants. In many Minnesota soils the major exchangeable bases are calcium and magnesium, sometimes constituting 90 percent or more of the total exchangeable cations. The fact that calcium and magnesium made up a relatively low percentage of the exchangeable cations in soils from the Brainerd milkshed indicated a low availability of these elements into plants. The low amounts of exchangeable calcium were also borne out by the low pH's of these soils. There appeared to be little in the data on exchangeable calcium and magnesium that would serve to distinguish high from low farms. If calcium (or magnesium) were substituting for strontium in farms with milk of low strontium-90 content, it was not apparent in the amounts of exchangeable calcium and magnesium in the soils.

The total stable strontium content of the soils was determined, since high stable strontium content might mean less absorption of strontium-90 by plants. What constitutes a "high" or "low" level of stable strontium is not known, since published values for stable strontium in soils are few in number. Fried and Heald (3), quoting Vinogradov, state that soils contain about 200 parts per million total strontium. This is much more than was found in the soils of the Brainerd milkshed, which might be too low to offer competition to absorption of strontium-90. However, the expected inverse relationship between total strontium and strontium-90 was not borne out by the data obtained in the soils analyses. Statistical comparisons were made of strontium and calcium concentrations in soils from high and low farms. There were significantly lower strontium concentrations in soils from low strontium-90 suppliers than from high suppliers, in both surface and subsurface soils, although this difference was more pronounced in surface soils. Calcium concentrations did not vary significantly in either surface or subsurface

soils between the 2 groups of farms.

Strontium-90 in Soils and Root Mats

The electrodialysis method of measuring strontium-90 in soils as described by Graham(4) seemed to offer economies in time and money; therefore, this method was selected as the most suitable for this study.

It became apparent after obtaining a number of values that concentrations were substantially below those obtained by extraction with 6N hydrochloric acid and those published by Alexander et al (2). However, a good estimate of the strontium-90 content of soils can be obtained by determining cesium-137 (5). Dr. P. F. Gustafson of the Argonne National Laboratory agreed to measure the cesium-137 content of the soil samples. Samples of soils collected in the fall had been depleted, but samples collected in the spring from identical sites were available and cesium-137 was determined on these. An examination of the data shows that amounts of cesium-137 in marsh areas (Barrows soil) were several times greater than in soils which were somewhat higher topographically. This would indicate either movement and accumulation of cesium-137 from higher to lower lying areas by water transport or greater retention of cesium-137 in the soils of greater organic matter content.

Whereas the average cesium-137 to strontium-90 ratio in soils was found to be 1.6 (5), Gustafson[1] suggested a value of 2 as perhaps being more appropriate. This ratio was used in calculating strontium-90 in this study. Since strontium-90 was calculated from cesium-137 values, the same pattern of deposition of these radionuclides would prevail. Amounts of strontium-90 per square mile approach those reported by Alexander et al (1) for other soils: viz., 59.1 mc/mi^2 to a depth of 6 inches at Mandan, North Dakota (1959). Data in this report were for surface soils only (the top 2 inches). Alexander et al (2) analyzed the 0-2 and 2-8 inch depths of some sites separately and found in general that the latter depth had from about half to two-thirds as much strontium-90 as the surface.

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[1] Personal communication from Dr. P. F. Gustafson, Argonne National Laboratory, Lemont, Illinois

Strontium-90 contents of 2 sites as determined by 6N hydrochloric acid extraction were available and there was good agreement between these values and those determined by calculation from cesium-137. These were too few samples to draw definite conclusions, but the good agreement between the results of the 2 methods of analysis was noteworthy.

One of the main purposes of this study was to trace, if possible, the source of strontium-90 which was contributing to the strontium-90 in the milk. There was no evidence from the few analyses available that the strontium-90 was higher in the soils on farms with higher strontium-90 in the milk, although marsh sites had significantly more strontium-90 than upland or lowland sites.

Root mats, which represent the surface vegetation plus roots separated from the mineral portion of the soil, were analyzed separately to determine the amount of strontium-90 in this soil fraction. Concentrations were found to be less than those in the soils from which the mats were extracted. It may be that a considerable, but undetermined, amount of radiostrontium may enter the food chain by lodging on aerial parts of plants. Russell (9) states that much of the strontium-90 absorption in established swards is due to "stem-base" absorption. In perennial grasses strontium-90 in roots may supply some of this element to above-ground parts. Nevertheless, the root mats contain only a small part of the store of strontium-90 in the soil, and when fallout becomes nil, in the case of a radioactive element with a half life of 28 years, a significantly active proportion of the element must ultimately be provided by the soil (10).

Calcium, Stable Strontium, and Strontium-90 in Vegetation

The stable strontium content of vegetation was uniform and there was no significant difference in amounts between farms with high and low strontium-90 contents in milk. Amounts of stable strontium in the soils were relatively uniform also. There was some variation in calcium content of vegetation from different soils, but the variation did not reflect the considerable differences in amounts of soil calcium. Strontium to calcium ratios were quite uniform in vegetation, but variable in the soils. Apparently plants were able to exercise some selection in the uptake of these ions and maintain a fairly constant ratio in their systems.

Calcium was significantly less in spring vegetation than in that taken in the fall, although high and low farms did not differ. It is likely that growth was greater in spring than in fall and the amount of calcium available was distributed through a greater quantity of plant material.

Strontium-90 was higher in spring vegetation than in fall samples, but, again, high and low farms did not differ. The fact that spring forage had more strontium-90 than fall materials may be due to greater fallout (more precipitation) in the spring (2). If the plant does accept strontium in place of calcium to some extent, then a deficiency in calcium in these soils (indicated by pH and exchangeable calcium) may contribute to an undetermined degree to the greater strontium-90 levels in spring vegetation.

The general concentration of strontium-90 in the vegetation could be considered to be low. An analysis of the vegetation species showed the grasses to be mostly mixtures of bluegrass and timothy (non-legumes). Vose and Koontz (10) studied the uptake of strontium and calcium from soils by grasses and legumes and concluded that every legume species takes up more strontium than any grass; within either the grasses or the legumes the variation in strontium content between the species and varieties was not marked; the amount of strontium taken up was directly related to the calcium taken up.

The data indicate that vegetation from marsh sites was significantly higher in strontium-90 than that from other sites, but that this was not peculiar to high or low farms. However, the fact that marsh vegetation had a higher strontium-90 concentration might be significant since farms with high strontium-90 milk had a greater proportion of the land in marsh (see appendix). If cattle grazed for longer periods on marshy areas, or if marsh hay made up a greater proportion of the diet of the cow, it is entirely possible, and indeed likely, that milk from these cows would contain more strontium-90. Information was not available, however, on which a conclusion could be reached regarding the proportion of feed coming from marsh, lowland, or upland areas.

The complementary relationship existing between strontium and calcium in plant nutrition is generally accepted. Uptake of strontium can be reduced by adequate supplies of calcium (8). Thus, a useful characterization of the strontium and calcium content of a plant is the strontium-90 to calcium ratio. Grass feeds with a high ratio would be expected to furnish more strontium to lactating animals than feeds of low ratio.

The data on strontium-90 to calcium ratios of vegetation show no clear-cut evidence that more strontium-90 was provided to animals in grass grown on farms with high strontium-90 milk. On the average, ratios for fall vegetation were higher for farms with low strontium-90 in milk; for spring vegetation, just the reverse was true. Ratios in spring vegetation were significantly higher than fall ratios. Almost without exception, vegetation on marsh sites had a higher strontium-90 to calcium ratio than on upland or lowland areas. This was true also for strontium-90 to strontium ratios. The most outstanding and consistent observation was the significantly higher amount of strontium-90 on the marsh sites, resulting in higher strontium-90 in the grasses and higner strontium-90 to calcium ratios.

CONCLUSIONS

The determination of such soil properties as organic matter, pH, magnesium, total calcium and stable strontium, cesium-137, and strontium-90 did not serve to distinguish between the survey farms with high or low strontium-90 concentrations in milk.

Total calcium, stable strontium and strontium-90 content, and strontium-90 to calcium and strontium-90 to strontium ratios of vegetation from the soil sampling sites did not distinguish between the survey farms.

Strontium-90 content of vegetation sampled in the spring was significantly higher than of fall vegetation from the same soil sampling sites.

Strontium-90 content of the mineral portion of the 0-2 inch soil layer was 5 to 30 times higher than in the associated root mat.

Strontium-90 and cesium-137 contents of soils and vegetation from marsh (Barrows) sites were significantly higher than those from lowland (Nokay) and upland (Brainerd) sites. The 2 survey farms with higher strontium-90 concentrations in milk had a greater proportion of the land in marsh. It was not possible to relate these facts to strontium-90 concentrations in milk, however, because it was not known how much of the diet of the cow was made up of grasses from the marsh areas.

SUMMARY

After consideration of all the data, there is still a question as to the causes for the observed differences in the concentrations of strontium-90 in the native grass hay.

The soils analyses do not explain these differences. Nor can they be explained on the basis of grass species, since the investigation of grass types growing in pasture did not show great differences between farms (this was only a visual survey and there may have been unascertained differences). The rain data do not shed any light on why these differences should occur, either. Thus, we must speculate.

It is noted from the soils data that the marsh areas showed higher strontium-90 concentrations in both the soils and vegetation than the better drained areas. Also, the farms producing the milk with the higher strontium-90 content had much more marshland available than those farms producing the milk with the lower strontium-90 content. Thus it is possible that the greater predominance of marshland and its use for pasture and hay might be the answer. This would indicate that the lower the land in terms of runoff, the higher the strontium-90 concentration in the soil and associated vegetation.

Such a conclusion implies that uptake from the soil is the dominant factor influencing strontium-90 concentrations in plants. This may or may not be the case, and some of the data from this study suggest that this is not the case.

From table 26 it can be seen that in most cases the strontium-90 to stable strontium ratio in the vegetation was several times that ratio in the associated soils. These higher strontium-90 to stable strontium ratios in plants might be the result of differences in the chemical availability of the strontium. This should be investigated further, but these ratios could also be the result of other factors. If it is assumed that the plant cannot discriminate between the 2 types of strontium (either chemically or physically), it must be concluded that a significant portion of the strontium-90 in the

plant came from a source other than uptake through the soil (e.g., surface deposition or foliar absorption).

Since the rain data did not show any great differences in total deposition of strontium-90, the differences in the grasses must be attributed to some other factor.

Assuming that uptake from the soil was small and deposition of strontium-90 per unit area by rain was essentially equal, one possibility for the differences in the plants could be differences in the mass productivity of the soil. In other words, the lower concentrations of strontium-90 would be found on the farms having the highest production in terms of weight per unit area simply by weight dilution.

The data from table 22 may be used to give some idea of productivity from the weight of sample collected per unit area. In this case, the quantity of grass obtained may in all probability depend more on the extent of grazing than the actual growth, but it does show there were differences in the quantities of grass available.

If from table 22 the yield in kilograms per square mile is calculated, it is found to be approximately twice as great on farms 2(L) and 10(L) as on farm 12(H). Unfortunately, the data are far too few and approximate to support anything but gross speculation.

In an attempt to obtain some data regarding surface deposition, several grass samples were rinsed in 5 percent nitric acid before analysis. Both the rinse water and the grass samples themselves were then analyzed for strontium-90 and calcium. The removal of strontium-90 ranged from 16 to 33 percent, with an average of 26 percent. The removal of calcium was insignificant. This indicates that a reasonable portion of the strontium-90 associated with grasses, if not just surface deposited, must be at least rather loosely bound.

An explanation of the differences in the concentrations of strontium-90 in the native grasses is beyond the scope of the data obtained, but this study should be helpful in determining what direction further study should take.

This study indicates that the major source of strontium-90 in milk is the native field grasses used for cattle feed. What this study has not shown is the means by which the strontium-90 becomes associated with these plants. There are several possible sources of strontium-90 in these grasses: uptake from the soil; foliar absorption or surface deposition

of fallout materials; surface deposition of soil dust; stem base absorption; and there may be others. Future studies should attempt to determine the relative importance of these various pathways, since control of strontium-90 in the plant will be possible only after the mechanisms of transport have been identified.

SUGGESTIONS FOR FURTHER STUDY

The Relative Importance of the Soil Contribution to Strontium-90 in Grasses

It was observed in this study that the strontium-90 content of different topographic soil locations differed significantly (marsh sites having the greatest amounts). A measure of the soil contribution to the field grasses could be obtained by growing these plants in sheltered locations in soils collected from the sites which showed differences in strontium-90 content. The effects of surface deposition or foliar absorption of fallout materials could be determined by equivalent and concurrent growing in unsheltered locations.

The Effects of the Deposition of Surface Soil Dust

Surface soils dust containing strontium-90 and deposited on the plant may be an important pathway of contamination. This aspect of contamination may be investigated by growing plants on selected soils in an enclosed chamber and spraying them with surface soil dust. Comparison of plants so treated with untreated plants grown under like condition should give some measure of dust retention and thus its importance as a pathway for contamination.

The Chemical Form of Stable Strontium and Strontium-90 in Soils

Relative to the uptake of strontium by plants is the nature of the compounds bearing this element. The availability of strontium to plants will be a function of the forms in which it exists. Strontium compounds in soil could be characterized by the quantities obtained in extracts varying in severity. These fractions could then be correlated to plant absorption to determine their relative contribution to strontium in the plant.

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APPENDIX I

Profile of Typical Brainerd Soil

Slope: 2 percent, westerly facing

Vegetation: mostly scattered oak

Description of the Profile

Horizon	Depth	Description
A_0	1-0 "	Rotted, well decomposed leaf mold of the multitype, some mixing with A_1 horizon.
A_1	0-3 "	Very dark brown sandy loam; weak, medium granular structure; friable; containing numerous fibrous roots.
A_{21}	3-6 "	Brown sandy loam; weakly granular structure; friable.
A_{22}	6-17"	Dark yellowish brown sandy loam; structureless; friable; some pebbles present.
B_1	17-21"	Dark brown sandy loam to sandy clay loam; mottles of light brown to pink; slightly cemented.
B_2	21-28"	Dark brown sandy clay loam mottled with pink and reddish yellow; weak, medium subangular blocky structure; moderately cemented.
C_1	28-33"	Dark brown sandy loam; some mottling as in B_2; moderately cemented.
C_2	33-48"	Dark brown sandy loam with some gravel; slightly cemented; occasional brown sandy clay areas.

Mechanical Analysis of the Profile

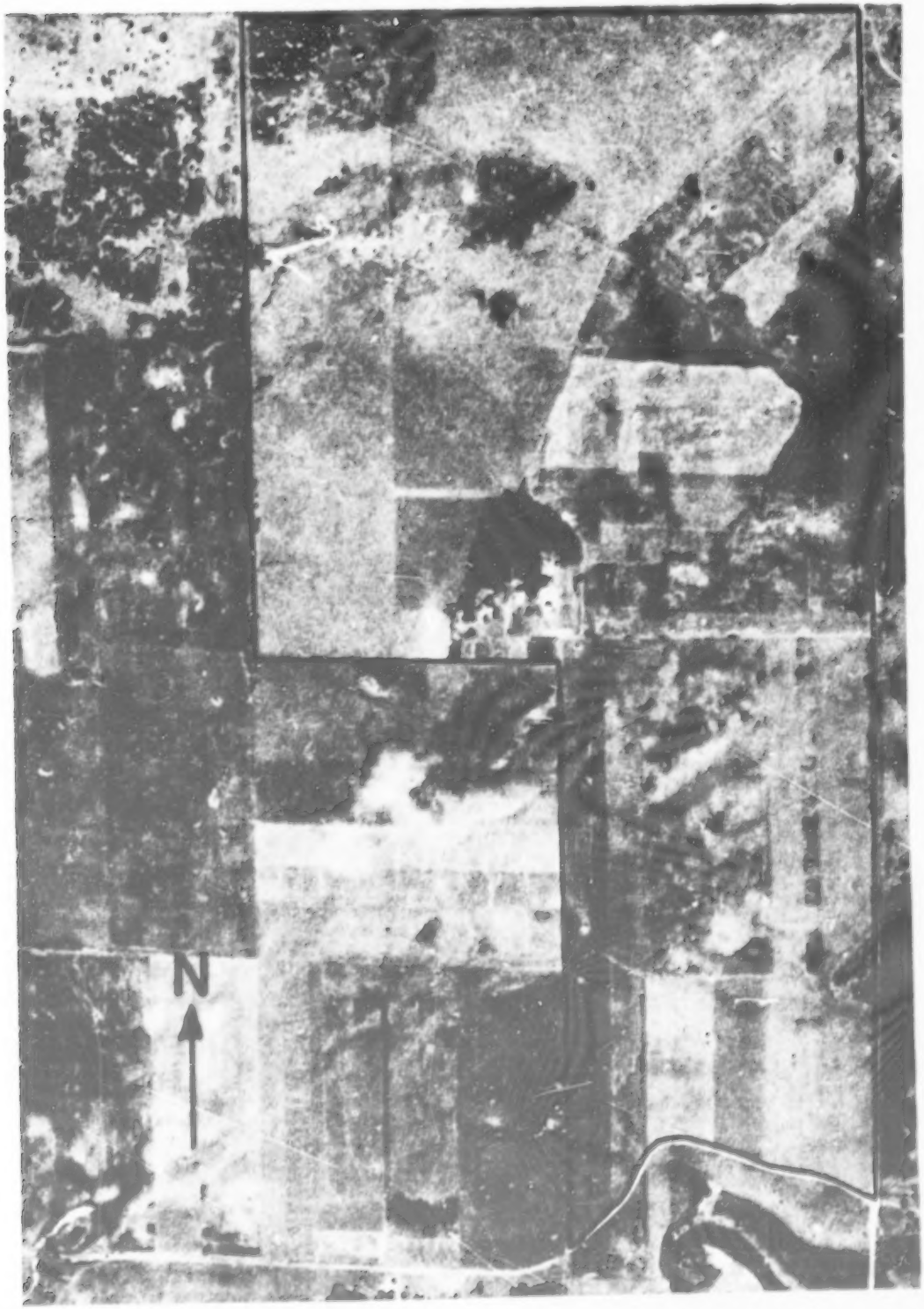
Horizon	Depth	Sand (>50 micron)	Silt (2-50 micron)	Clay (<2 micron)
A_0	1-0 "	*	*	*
A_1	0-3 "	59.0	33.2	7.8
A_2	3-17"	67.1	27.1	5.8
B_1	17-21"	70.1	18.3	11.6
B_2	21-28"	71.5	17.5	11.0
C_1	28-33"	75.8	16.3	7.9
C_2	33-48"	71.7	18.2	11.1

* Not determined.

APPENDIX II

Aerial Photographs of Farms

Aerial Photograph of Farm 2(L)



Aerial Photograph of Farm 10(L)

Aerial Photograph of Farm 12(H)

Aerial Photograph of Farm 14(H)

APPENDIX III

Area of Each Soil Series Found on the Brainerd Milkshed Farms

Soil series and slope grouping	Farm 2(L)		Farm 10(L)		Farm 12(H)		Farm 14(H)	
	Acres	Percent of total	Acres	Percent of total	Acres	Percent of total	Acres	Percent of total
Chetek sandy loam								
0-2%			15	5				
2-7%			2	1				
7-13%			8	3				
Burkhardt sandy loam			44	16	6	3		
Halder loam			14	5				
Warman loam			4	1				
Menahga loamy sand and sand			26	9				
Lino loamy sand			1	0				
Brainerd sandy loam								
0-2%	24	8	10	4	28	14	80	50
2-7%	98	34	103	37	25	12	12	8
7-13%			7	2				
Nokay sandy loam								
0-2%	121	41	6	2	55	28	31	20
2-7%	21	7						
Barrows loam and sandy loam	32	10	2	1	2	1	4	2
Peat								
Less than 36"			17	6				
Deeper than 36"			21	8	56	28		
Marsh					28	14	33	20
Total acreage	296		280		200		160	
Percent acreage less than 2% slope		59		57		87		90
Percent acreage more than 2% slope		41		43		13		10

Note: On farm 10(L) the Chetek, Burkhardt, Halder, Warman, Menahga, and Lino soils did not lie in the sample areas.

APPENDIX IV

The Use of Electrodialysis in Strontium-90 Extraction

INTRODUCTION

In an effort to measure the soil nutrients available to plants, a number of extracting procedures have been proposed, including distilled water, weak acids, and neutral salts. In 1926 Bradfield (1) proposed electrodialysis as a means of extracting the available basic ions from the soil colloids. In this procedure a direct current is applied to the soil mass which is contained in a semi-permeable membrane (commonly, a Visking membrane). The electrodes may be placed in a 3-cell compartment containing, e.g., an inner electrode of platinum and 2 outer electrodes of platinum or stainless steel.

The work of Marshall (5) and others has demonstrated that ions are held on the colloids with variable binding energies. Some ions are held so firmly as to be essentially unavailable to plant roots. However, the "impressed" energy in electrodialysis is considered sufficient to extract all the available nutrients. In fact, most recent soil research suggests that electrodialysis is too severe an extracting procedure and neutral salts are more commonly used. In a study of strontium-90 and cesium-137 uptake by plants, Graham (2) reported higher correlated uptake of strontium-90 extracted by water than by strong acid treatment.

In this study electrodialysis was used because it was felt that any error in the measurement of strontium-90 available to plants should be on the excess side. A second reason for using this method was to avoid "loading" the soil extract with a large number of extraneous ions, particularly iron, which would be extracted by strong acid. The work of Hamada et al (4) indicated that the problem of scavenging extraneous ions is laborious and subject to manipulative error. Thirdly, the work of Graham (3) indicated that, with suitable experimental arrangements, electrodialysis of a sufficient amount of soil, of the order of 500 grams, could be accomplished in several hours.

The cell construction and the power source used were

essentially as outlined by Graham (3).

For the separation of calcium from other ions in the dialysate an ion exchange column was employed. The calcium was chelated with EDTA at about pH 10. The pH was lowered to 5.5, releasing strontium, and the solution poured over Dowex 50 resin. Alkaline earths, magnesium, some remaining calcium, and yttrium-90 were removed from the column by a mixture of EDTA (0.75 percent) and citric acid (1.0 percent). The columns were then sent to the Minnesota Department of Health laboratories where the strontium-90 was stripped from the resin with 6N hydrochloric acid and purified, using appropriate steps in the general procedure described elsewhere in these appendixes.

The amount of strontium-90 removed from soils by this method has been found to be less than that recovered by 6N hydrochloric acid extraction. According to Hamada et al (4) the strong hydrochloric acid extraction recovers about 80 percent of the total strontium-90 using sodium carbonate fusion analysis as a standard comparison. Graham (3) states that electrodialysis will remove strontium-90 which exists in salt form or as an exchangeable ion. Therefore, it appears that a significant amount of strontium-90 exists in some mineral or mineral-organic form which is not recovered by electrodialysis and would apparently not be immediately available to plants. It is quite possible that, whatever the chemical nature of "bound" strontium-90 may be, it will become available to plants as a result of subsequent weathering action in the soil environment. It is also possible that a portion of the strontium-90 will leach downward and beyond the reach of growing plant roots.

METHOD OF ANALYSIS

Apparatus

Electrodialysis cells, constructed from Plexiglass
D-c source, 250 watt capacity, constructed
Ion exchange columns, 20 cm length and 1.6 cm inside diameter
4-inch Pyrex funnels with rubber stopper fittings
Glass electrode pH meter
Stirring arrangement

Reagents

Dowex 50 W-X12 type resin, 50 to 100 mesh
NaOH solutions: 5% and 1N
HCl solutions: 5%, 1N, and 6N
H_2O_2 solution: 30%
NaCl solution: 5%
Indicator: 1.5 gm Erichrome black T and 4.5 gm hydroxylamine hydrochloride dissolved in 100 ml ethyl alcohol; filtered
EDTA, tetra sodium salt, powder, technical
EDTA plus citric acid solution: 1% citric acid monohydrate and 0.75% EDTA solution adjusted to pH of 5.0 with 5% NaOH and HCl

Procedure

A. Extraction by electrodialysis

1. 100 grams of air dried soil which had been passed through a 2 mm sieve were placed in the anode chamber.
2. The cell was filled with deionized water.
3. The voltage was maintained at about 100 volts d-c and the current at about 2 amperes through the 5 cells connected in series.
4. The liquid was removed from the cathode chamber at 0, 2, 4, and 10 hours after initiation of the current. Each time, the cell was refilled with deionized water.
5. The basicity of these extracts was determined by titration with 1N HCl to the phenolphthalein end point. These extractions furnished electrodialysis-extractable total bases.

B. Separation of calcium and magnesium from strontium; chelation of calcium and magnesium

1. The volume was reduced to 500 cc by slow evaporation.
2. A small amount of 30% hydrogen peroxide was added.
3. The solution was heated for a time to destroy any organic matter.
4. Enough EDTA powder was added to chelate the calcium and magnesium. This was about 1 gm

per 3.2 milliequivalents of electrodialysis-extractable total bases.

5. The solution was stirred and adjusted to pH 10 by adding 5% NaOH.
6. Complete chelation of the calcium and magnesium was checked by adding a few drops of Erichrome black T indicator. If the solution was pink, additional chelate was needed.
7. PH of solution was reduced to 5.5 with 5% HCl.

C. Ion exchange separation of strontium from calcium and magnesium

1. The resin was activated with several washings of 5% HCl alternated with 5% NaCl, then with 5% NaCl, and finally washed with deionized water until free of chloride ion.
2. The chelated solution was passed through the resin column at a flow rate of about 8 ml per minute per cm^2.
3. 500 ml of EDTA plus citric acid solution were passed through the column.
4. 100 ml of deionized H_2O were passed through and the effluent discarded.

DETERMINATION OF THE AMOUNT OF STRONTIUM-90

The resin columns were delivered to the Division of Environmental Health, Minnesota Department of Health, for assay of strontium-90. The results of the analyses are given in table A, which also includes for comparison the amounts of strontium-90 determined from the cesium-137 analyses. The results are expressed in micromicrocuries per kilogram.

The strontium-90 content as determined by electrodialysis was, in all cases, less than that calculated from the cesium-137 determinations, varying from 9 to 64 percent of the latter values. On the average, the electrodialysis results are about one-third of the values by cesium-137 analyses. The range of findings for both analyses show a 4- to 5-fold variation comparing low to high values.

Assuming that strontium-90 content as determined by cesium-137 analysis represents a true estimate of the strontium-90, the following possibilities may explain the electrodialysis results:

(1) electrodialysis did not extract all the strontium-90 in the soil; and

(2) some strontium-90 was lost in the process of separation on the resin column or in later handling.

Table A.--Comparison of Strontium-90 Content as Determined by Electrodialysis to That Determined by Cesium-137 Analysis

Soil number	Sr^{90} content (μμc/kg)		Percent electrodialysis of Cs result
	Electrodialysis	Cs^{137} analysis	
19-L-U-S	106	168	63
21-L-M-S	76	868	9
23-L-M-S	308	821	38
25-L-L-S	91	-	-
27-L-M-S	54	310	17
29-H-U-S	112	177	64
31-H-L-S	60	312	19
33-H-M-S	59	-	-
35-H-M-S	113	723	16
37-H-L-S	68	200	34
Average			32

In regard to the first possibility, it should be noted (as indicated in the introduction to this appendix) that the amount of bases removed by electrodialysis is an approximation of those ions which may be available to plants ("available" as previously defined). In table B the bases removed by electrodialysis are compared to the theoretical total amount of bases, estimated by complete replacement. The results are given in milliequivalents per 100 grams. It appears that, on the average, 57 percent of the total replaceable bases in the soil were removed in the dialysate.

Therefore, one conclusion might be that two-thirds of the strontium-90 not extracted by electrodialysis remains in the 40 to 45 percent of the bases not removed (table A). The work of Roberts and Menzel (6) on surface soil in the coastal plane of North Carolina suggests that from 5 to 50 percent of the strontium-90 is in a form requiring at least treatment by heated 4N HCl for removal. Their work further indicates that only 8 to 18 percent of the exchangeable strontium-90 was taken up by a legume (cowpeas) in one year's growth.

Thus, the values obtained by electrodialysis, while not giving a true estimate of the total strontium-90 in the soil, may correlate more closely with uptake by forage and other plants.

Table B.--Titratable Bases in the Electrodialysate and Total Base Exchange Capacity of the Surface Soil in the Brainerd Milkshed Area

Soil number	Titratable bases[1] (me/100 gm)	Base exchange capacity[2] (me/100 gm)	Percent bases of capacity
19-L-U-S	10	18	55
21-L-M-S	18	28	64
23-L-M-S	13	-	-
25-L-L-S	8	27	30
27-L-U-S	22	25	88
29-H-U-S	5	14	36
31-L-H-S	18	26	69
33-H-M-S	21	-	-
35-H-M-S	20	35	57
37-H-L-S	22	-	-
Average			57

[1]Determined by HCl titration.
[2]Determined by amperometric titration of silver acetate.

The possibility that some strontium-90 was lost in the process of separation exists. Hamada et al (4) reported that, even with careful scavenging of interfering ions, 10 to 15 percent of the strontium-90 may be lost in precipitation. Some strontium-90 analyses were made on electrodialysates handled in 2 ways: (1) as described in the previous section, Method of Analysis; and (2) by using successive oxalic acid precipitations to remove yttrium and rare earth interferences. Table C gives the comparison of results. These data suggest that if loss of strontium-90 occurs, it may be slightly less by the resin separation than by separation with successive precipitations.

Table C.--Amounts of Strontium-90 Determined on Electrodialyses Extracts by 2 Procedures

Soil number	Sample size (grams)	Strontium-90 (μμc/kg)	
		Resin separation	Oxalic acid precipitation
18-L-U-S	600	73 ± 5	52 ± 8
[a]19-L-U-S	360	150 ± 6	144 ± 18
[b]31-H-L-S	297	138 ± 7	168 ± 23
[c]37-H-L-S	400	128 ± 5	103 ± 12

[a] 50 μμc Sr^{90} added/kg.
[b] 60 μμc Sr^{90} added/kg.
[c] 45 μμc Sr^{90} added/kg.

Thus, it appears that the lower amounts of strontium-90 determined by electrodialyses, as compared to those calculated from cesium-137 values, were related to the method of extraction from the soil. Further investigation is indicated to establish the degree of correlation between electrodialyzable

strontium-90 and plant uptake of this ion.

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APPENDIX V

Determination of Strontium-90 in Milk*

GENERAL METHOD

Reagents

1. Strontium nitrate solution. Dissolve 9.644 gm of $Sr(NO_3)_2$ in distilled water and dilute to 1 liter. One ml of this solution contains 4 mg of Sr.
2. Nitric acid, 1:1. Dilute 500 ml of concentrated HNO_3 with an equal volume of distilled water.
3. Nitric acid, 5%. Dilute 50 ml of concentrated HNO_3 to 1 liter with distilled water.
4. Oxalic acid, 10%. Dissolve 100 gm oxalic acid ($H_2C_2O_4 \cdot 2H_2O$) in 1 liter of distilled water on a steam bath.
5. Ammonium hydroxide, 1:5. Dilute 200 ml of concentrated NH_4OH to 1 liter with distilled water.
6. Wetting agent, dilute. Dilute 1 drop of Sterox with 10 ml of distilled water.
7. Yttrium solution, 2 mg/ml. Dissolve 8.0 gm $Y(NO_3)_3 \cdot 4H_2O$ in 1 liter of distilled water.
8. Ammonium chloride solution, 0.2 gm/ml. Dissolve 200 gm NH_4Cl in distilled water and dilute to 1 liter.
9. Sodium hydroxide, 10%. Dissolve 10 gm NaOH in distilled water and dilute to 100 ml.
10. Methyl orange indicator, 0.1%. Dissolve 0.1 gm of the sodium salt of p-dimethylaminoazobenzene-sulfonic acid in 100 ml distilled water.
11. Hydrochloric acid, 1 normal. Dilute 8.3 ml of concentrated HCl to 100 ml with distilled water.
12. Barium chloride solution, 0.1 N. Dissolve 12.0 gm $BaCl_2$ in 1 liter of distilled water.

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* From the Minnesota Department of Health, Section of Engineering Laboratories.

13. Ammonium dichromate solution, 0.1 M. Dissolve 25.2 gm $(NH_4)_2Cr_2O_7$ in distilled water and dilute to 1 liter.
14. Acetate buffer. Dissolve 154 gm $NH_4 \cdot C_2H_3O_2$ and 60 ml glacial acetic acid in distilled water and dilute to 500 ml.
15. Sodium carbonate, 3N. Dissolve 160 gm Na_2CO_3 in distilled water and dilute to 1 liter.
16. Hydrochloric acid, 6N. Dilute 1 volume of concentrated HCl with an equal volume of distilled water.
17. Gelatin solution, 0.2%. To 50 ml of distilled water add 0.1 gm gelatin (1 scoopful). Heat to near boiling on a steam bath to dissolve.

Procedure

1. Add 800 ml of a milk sample (whole or skim milk) to a 1 liter evaporating dish (pyrex) and add 20 mg of Sr^{++} as $Sr(NO_3)_2$.
2. Dry the milk on a steam bath, then ash at 450° C for at least 16 hours.
3. Cool to room temperature and add 40 ml of 1:1 HNO_3, washing down the sides of the dish with the acid solution. Evaporate to dryness and ash for 1 hour at 450° C. Cool, add 20-30 ml of 1:1 HNO_3 and stir.
4. Evaporate to dryness on a steam bath.
5. Add 30 ml of a 5% HNO_3 solution to the evaporating dish and digest on a steam bath for 20 minutes to dissolve the residue. The evaporating dish may have given up some silica flakes which will appear in the ashed residue; these will not dissolve.
6. Filter the solution of the residue through a glass fiber mat or a filter paper mat. Rinse the evaporating dish with two 15 ml portions of the 5% HNO_3 solution, filter and combine the rinse with the filtered solution of the residue.
7. Transfer the filtrate to a 1 liter beaker, dilute to about 500 ml and heat to near boiling.
8. Add 60 ml of 10% oxalic acid solution, stir, and digest on a steam bath for about 20 minutes.
9. While still hot, add dropwise and with constant stirring dilute NH_4OH solution until a pH of 4.0 is reached.
10. Add about 10 drops of dilute wetting agent solution, stir, and let stand overnight.
11. Decant or draw off the supernatant liquid and transfer the precipitate quantitatively to a 50 ml centrifuge

tube; centrifuge, decant, and save the supernatant for cesium determination.

12. Add about 25 ml of distilled water to the centrifuge tube, mix the precipitate with water, centrifuge, decant, and discard the supernatant.
13. Repeat step 12.
14. Add 5 ml of concentrated HNO_3 to the centrifuge tube; do not stir or mix. Add 10 ml of 30% H_2O_2; do not stir or mix as reaction will proceed too rapidly if contents are mixed before placing on the steam bath.
15. Place a no. 19 glass stopper (flat top) in the top of the centrifuge tube, then heat the centrifuge tube in a hot water bath or slow steam bath. If reaction proceeds too fast the sample may be lost. If and when the froth from the reaction approaches the top of the centrifuge tube, remove the tube from the bath and place in a beaker of cold water. When the reaction has slowed up considerably, return the tube to the steam bath and observe from time to time, making sure to avoid loss of sample caused by too rapid a reaction rate.
16. When the reaction has subsided to a low evolution of gas, swirl the tube gently to mix the contents.

    Note: 10 ml of 30% H_2O_2 is not usually enough to destroy all of the oxalate. However, before adding any more H_2O_2, be sure that the reaction of the first 10 ml is quite complete. One hour is usually sufficient time.

    If on swirling the tube there is no great increase in the evolution of gas, then the second and final 10 ml of H_2O_2 should be added. Again, do not mix the contents after the addition of H_2O_2; let the evolution of the gas do the mixing.
17. When the evolution of gas is no longer evident, remove the glass stopper and evaporate the contents of the centrifuge tube to near dryness on the steam bath.
18. Add 10 ml of yttrium solution, 2.5 ml of ammonium chloride solution, 1 ml of barium chloride solution, 1 ml of ammonium dichromate solution, and 4 drops of wetting agent to the centrifuge tube. If considerable acid is present, partially neutralize with concentrated NH_4OH to a point just before a precipitate persists.
19. Dilute to about 40 ml and heat on a steam bath.
20. Slowly raise the pH of the solution, using a pH meter, to 8.0 with dilute NH_4OH.
21. Digest the sample for 20 minutes on the steam bath.

22. Add 2.5 ml of the yttrium solution and 1 ml of the barium chloride solution. Stir and check the pH. Digest for another 20 minutes.
23. Dilute to 50 ml and cool. Centrifuge for 20 minutes.
24. Carefully and completely decant the supernatant into a 125 ml Erlenmeyer flask. Allow the centrifuge tube to drain for at least a half minute. Discard the precipitate.
25. Add sufficient 6N HCl (about 1 ml) to lower the pH to 1.5 or lower and add 2 to 3 ml of ethyl alcohol.
26. Add 2.5 ml of yttrium solution and allow the sample to stand about 2 weeks for yttrium-90 ingrowth.
27. After 2 weeks ingrowth of yttrium-90, evaporate the solution to a small volume (about 20 ml) on a steam bath.
28. Transfer the solution quantitatively to a 50 ml centrifuge tube, dilute to 30 ml, heat on a steam bath for 5 to 10 minutes, and add 2 drops of dilute wetting agent.

Note: Steps no. 29 through 37 should be completed the same day.

29. While the solution is hot, adjust the pH to the methyl orange end point (yellow) using concentrated NH_4OH, then add 3 drops of concentrated NH_4OH, stir, digest for 10 minutes, add 2.5 ml more of the yttrium solution, stir, digest for another 10 minutes, and centrifuge. Note time; this time is the end of the ingrowth of yttrium-90. Both the precipitate and supernatant are to be saved.
30. Decant the supernatant solution to a 100 ml volumetric flask. Wash down the sides of the centrifuge tube and stir the precipitate with 10 ml of a dilute NH_4OH solution (1 drop of concentrated NH_4OH in 50 ml of distilled water), centrifuge, and add the wash solution to the 100 ml volumetric flask. Wash with another 10 ml of dilute NH_4OH solution and again add this wash to the 100 ml volumetric flask. The combined supernatant and wash solution is to be used for the assay of calcium and for the determination of strontium recovery. This solution should be acidified with 1.0 ml of 6N HCl after the withdrawal of aliquots for strontium and calcium assay. The solution may be allowed to produce more yttrium-90 for a later count.
31. Add sufficient 6N HCl (5 drops) with stirring to dissolve the precipitate in the centrifuge tube, dilute to 30 ml, while heating on a steam bath, add 5 ml of a

10% oxalic acid solution, stir, add NH_4OH to the methyl orange end point (pH = 4.0), add 4 drops of the dilute wetting agent, digest sample for 10 minutes, and, finally, cool the mixture.

32. Prepare a Millipore filter on the Millipore filter assembly, wetting the filter thoroughly before closing the assembly, and mount the assembly on a suction flask. Do not apply suction until just before the contents of the centrifuge tube are to be added. It is important to avoid drawing air through the Millipore filter because air filtered through the Millipore filter will leave activity (daughter products of radon) on the filter, thus giving erroneous results when counting the yttrium-90.
33. Suspend the yttrium precipitate in the centrifuge tube by stirring, then apply suction to the suction flask and promptly add the contents of the centrifuge tube to the filter assembly. Rinse the centrifuge tube, quantitatively transferring the yttrium precipitate onto the Millipore filter. Be careful to cut off the suction any time the filter appears to be drained, and also to turn on the suction again just before any rinse solution is added to the filter (otherwise, solids on the filter may be disturbed).
34. Add 2.0 ml of fresh 0.2% gelatin solution to about 20 ml of distilled water and add this solution to the Millipore filter assembly (after turning on the suction again). When the filter appears drained, cut off the suction.
35. Prepare an aluminum planchet (2" diameter), properly identified on the bottom, with a thin film of high temperature silicone grease spread uniformly over the inside bottom of the planchet.
36. Carefully transfer the wet Millipore filter to the planchet being careful to center the filter in the planchet. Dry the filter with a heat lamp about 1 foot distance from the filter.
37. Count promptly after drying. Note counting time. A low background beta anti-coincidence counter should be used for counting. Count for 2 hours.

SHORT METHOD

(Phosphate Precipitation)

Reagents for this method are the same as those listed for the general determination.

Follow the regular procedure through step 6. At this point, the sample should be at least 2 weeks old to insure strontium-90 - yttrium-90 equilibrium. The sample may be aged at any stage in the procedure.

Continue the analysis as follows:

7. Transfer the filtrate to a 1 liter beaker, add 20 mg of yttrium carrier, and dilute to about 500 ml. Heat to near boiling.
8. Slowly add 1:5 NH_4OH with constant stirring (magnetic stirrer) while the sample is still hot until the pH has been raised to 2.6.
9. Cool to room temperature and filter through a membrane filter. Pass 10 ml of water containing 2 mg of gelatin through the filter.

Continue with step 35 of the general procedure.
Recount after 2 weeks as a check on yttrium-90 purity.

APPENDIX VI

Levels of Cesium-137 Found in Milk

Date of collection		μμc Cs^{137} per liter			
		Farm 2(L)	Farm 10(L)	Farm 12(H)	Farm 14(H)
1959					
November	17		41	40	71
	30	16	34	57	56
December	16	33	27		
	28	41			41
1960				110	58
January	11	29		140	120
	25	56	41	98	60
February	9	23	29	81	100
	26	45	45	64	95
March	7	46	40	75	110
	21	42	28		90
April	4	26	34	55	44
	18	36	30	60	61
May	2	50	49	49	130
	16	78	21		120
	31	71	52	146	105
June	13	71	78	146	141
	27	79	48		

APPENDIX VII

List of Plants in Pastures, November 1, 1959

Scientific name	Common name	Farm 10(L)			Farm 2(L)				Farm 12(H)		Farm 14(H)	
		100-acre pasture	Cultivated	Marsh	Main pasture	Weed patch	Back pasture	Marsh pasture	Main pasture	Slough	Main pasture	Wooded pasture
Achillea millefolium	Yarrow			X			X		XX		X	X
Agropyron repens	Quackgrass	X									X	
Alisma triviale	Water plantain									XXX		
Amaranthus retroflexus	Pigweed										X	
Ambrosia artemisiifolia	Ragweed	X			XX							X
Antennaria sp.	Pussy toes	X							XXXX		XX	X
Betula papyrifera	Paper birch								borders X			
Betula pumila	Bog birch								borders X			
Chenopodium album	Lambsquarters					XX						
Cirsium arvense	Canada thistle						XXX		X		X	XX
Cirsium vulgare	Bull thistle								X			
Corylus americana	Hazel brush						X					
Festuca sp.	Fescue grass	XX					XXX					
Iris versicolor	Iris											X
Linaria vulgaris	Toadflax	X				XX						X
Lycopodium sp.	Club moss							XX			X	
Medicago sativa	Alfalfa				X							
Panicum sp.	Panic grass	XX		XX	X		X		XXX	XXX	X	X
Phleum pratense	Timothy	XXX		XXXX		XXX	X		XX		X	
Plantago major	Plantain				XXX				X		X	
Poa pratense	Blue grass	XXXX		X	XXXX		XXXX	XXX	XXXX		XXXX	XXXX

List of Plants in Pastures, November 1, 1959, cont'd.

Scientific name	Common name	Farm 10(L)			Farm 2(L)				Farm 12(H)		Farm 14(H)	
		100-acre pasture	Cultivated	Marsh	Main pasture	Weed patch	Back pasture	Marsh pasture	Main pasture	Slough	Main pasture	Wooded pasture
Polygonum punctatum	Smartweed							X				XX
Populus sp.	Aspen								X			
Potentilla argentea	Silvery cinquefoil	X			X			XX				
Potentilla norvegica	Rough cinquefoil	X				XXXX			X		X	
Quercus alba	White oak											X islands
Rorippa islandica	Yellow cress					X						
Rosa arkansana	Wild rose						X islands					
Rumex acetosella	Sheep sorrel					XX		XX				
Salix sp.	Willow			XX				XXX	Island & border X			
Secale cerea	Rye		XXXX									
Setaria glauca	Pigeon grass				XX							
Setaria italica	German millet		XXXX									
Silene sp.	Catchfly				X							
Sedges	Sedges			XXX				XXXX	X	XXX	borders X	
Taraxacum officinale	Dandelion				X				X		X	
Trifolium pratense	Red clover	X			XX				XX			
Trifolium repens	White clover				XXX				X			
Verbascum thapsus	Mullein		X			X	X					

List of Plants in Pastures, June 30, 1960

Scientific name	Common name	Farm 10(L)	Farm 2(L)	Farm 12(H)	Farm 14(H)
Achillea millefolium	Yarrow	XX	X	X	T
Agropyron repens	Quackgrass	X	T	0	T
Agrostic alba	Redtop	XXX	X	T	X
Ambrusia artemisiifolia	Ragweed	0	T	0	T
Antennaria sp.	Pussy toes	X	T	XXX	XX
Betula papyrifera	Paper birch	0	0	T	0
Betula pumila	Bog birch	0	0	T	0
Chenopodium album	Lambsquarters	0	0	0	X
Cirsium arvense	Canada thistle	X	X	T	X
Cirsium vulgare	Bull thistle	0	0	X	T
Corylus americana	Hazel brush	T	X	X	0
Fragaria virginiana	Wild strawberry	X	0	X	T
Iris versicolor	Iris	0	0	0	T
Linaria vulgaris	Toadflax	X	X	0	0
Lycopodium sp.	Club moss	0	0	XX	X
Panicum sp.	Panic grass	?	0	0	0
Phleum pratense	Timothy	XX	XXX	XX	XX
Plantago major	Plantain	0	X	0	0
Poa pratense	Blue grass	XXXX	XXXX	XXXX	XXXX
Polygonum sp.	Smartweed	0	T	0	X
Populus sp.	Aspen	T	0	T	0
Potentilla argentea	Silvery cinquefoil	X	T	0	0
Potentilla norvegica	Rough cinquefoil	X	0	0	X
Quercus alba	White oak	0	XX	0	0
Rorippa islandica	Yellow cress	0	T	0	0
Rosa arkansana	Wild rose	0	0	T	0
Rumex acetosella	Sheep sorrel	X	0	T	0
Salix sp.	Willow	T	0	T	0
Sedges	Sedges	X	XXX	XX	XX
Taraxacum sp.	Dandelion	0	X	T	X
Trifolium pratense	Red clover	X	T	0	0
Trifolium repens	White clover	XXX	XX	XX	XX
Verbascum thapsus	Mullein	0	X	0	0
Vicia sp.	Vetch	X	T	T	T